

Supporting Information for

Tracking regional CH₄ emissions through collocated air pollution measurement: a pilot application and robustness analysis in China

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This PDF file includes:

Figures S1 to S8
Tables S1 to S3
SI References

Supporting Figures

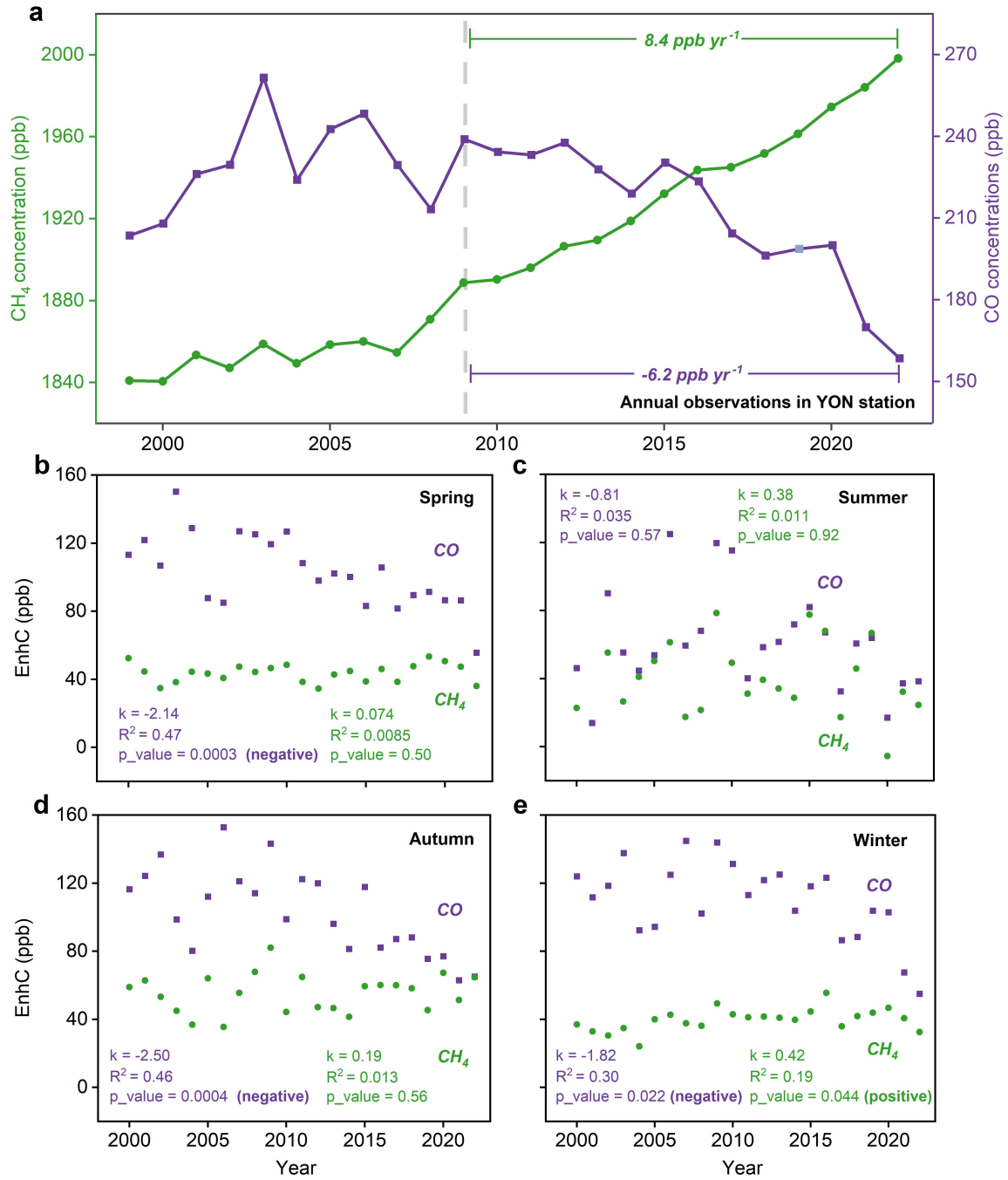


Fig. S1. The annual and seasonal variations of CO and CH₄ concentrations from 2000 to 2021. (a) Annual average concentrations of CO and CH₄ as measured at the YON station. (b–e) Seasonal *EnhCs* of CO and CH₄, were calculated by subtracting the observations at the MNM background station from those at the YON station. The *EnhCs* of CH₄ show significant decreasing trends in spring, autumn, and winter, while the *EnhCs* of CO indicate a significant increasing trend in winter.

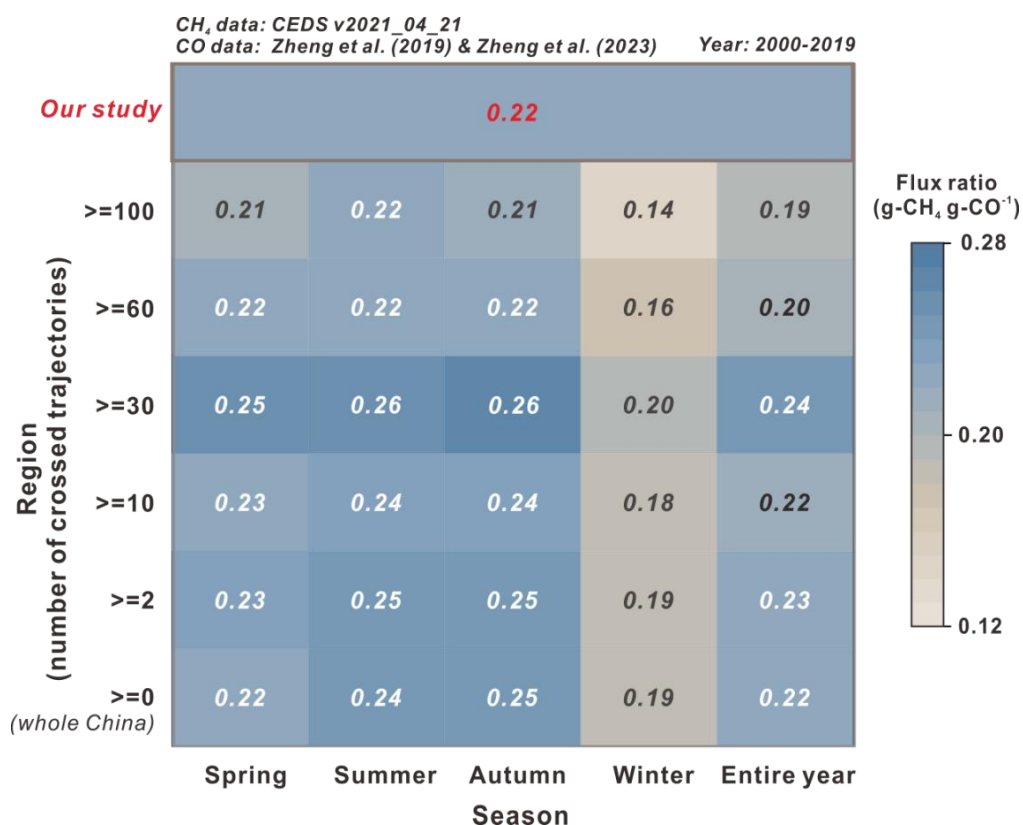


Fig. S2. Comparisons of CH₄-to-CO flux ratios under different CH₄ data and across various spatio-temporal scales. The color and value of each pixel represent the mean emission ratio for the period 2000-2019. The ratios in the lower section of the figure are determined using CH₄ data from CEDS (v2021_04_21)(1) and our CO inversion results (2, 3), while the ratio in the top section is derived from our study. Values in the bottom are calculated across different temporal partitions—spring, summer, autumn, winter, and the entire year—as well as across various spatial distributions, categorized by the number of trajectory crossings within a 0.5° grid over China, with thresholds of ≥0, ≥2, ≥10, ≥30, ≥60, and ≥100 crossings.

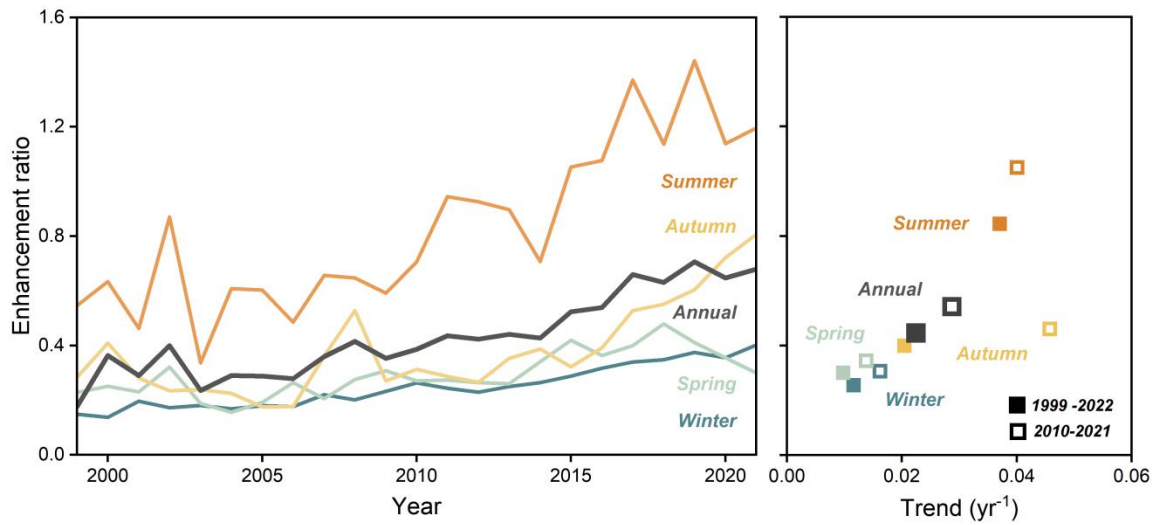


Fig. S3. Seasonal and annual *EnhRs* and their trends from 1999 to 2021. The left panel shows the seasonal and annual variations in *EnhRs* from 1999 to 2021. The right panel illustrates the corresponding trends for each season and the year, with trend comparisons for the periods 1999–2022 and 2010–2021, respectively.

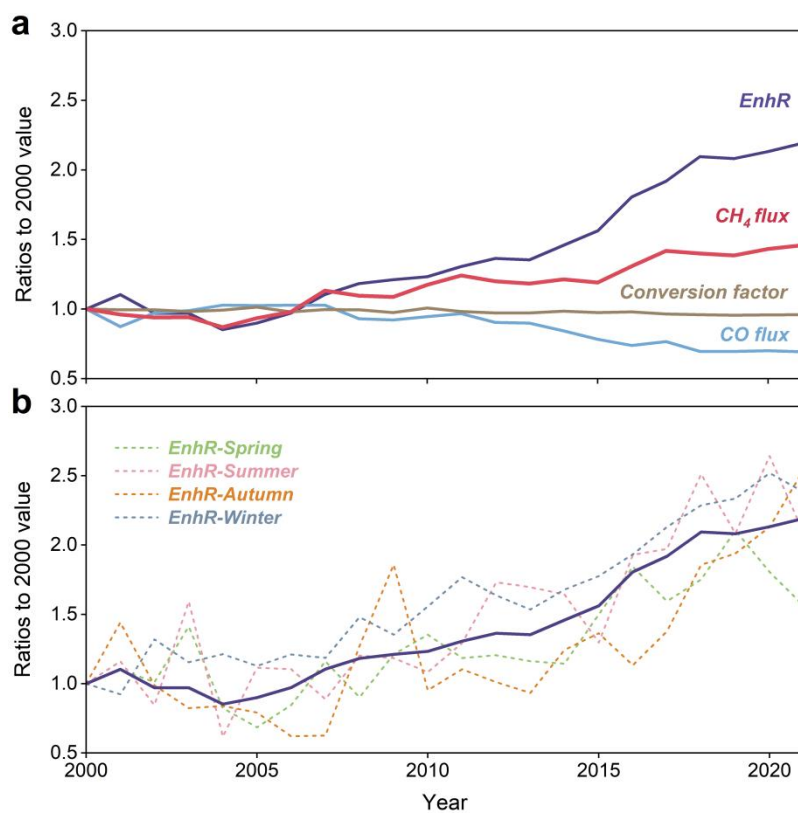


Fig. S4. Ratios of key parameters relative to their values in the year 2000, spanning from 2000 to 2021. (a) Ratios of parameters including *EnhR*, conversion factor, CO flux, and CH_4 flux to their respective values in 2000. **(b)** Ratios of seasonal *EnhRs* to their values in 2000.

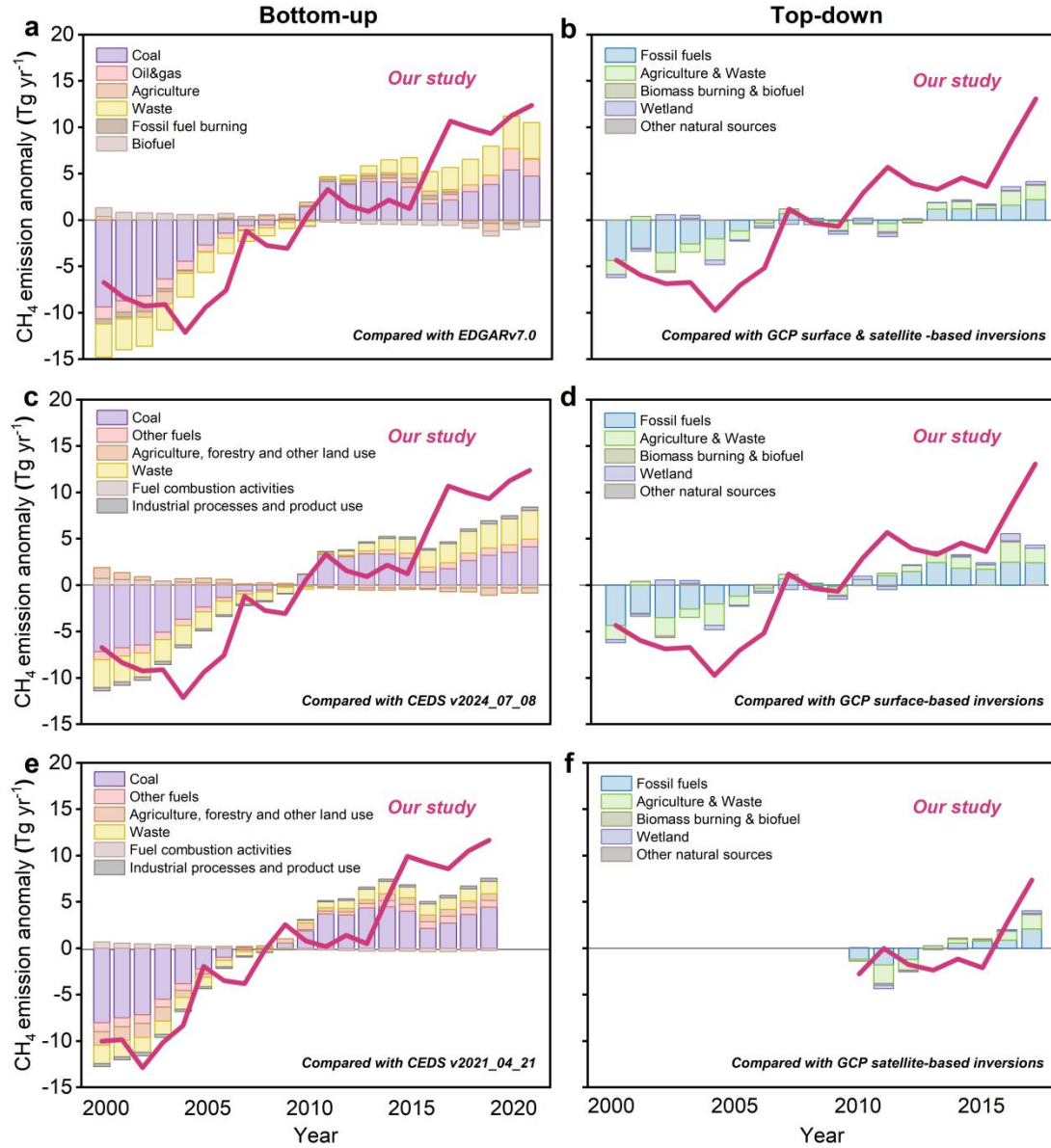


Fig. S5. CH₄ emission anomalies from our study compared with other independent bottom-up (BU) and top-down (TD) results containing detailed sectoral data. The anomalies are calculated as deviations from the average values over the study period. **(a)**, **(c)**, and **(e)** compare our emission anomalies with bottom-up inventories: EDGARv7.0 (https://edgar.jrc.ec.europa.eu/dataset_ghg70), CEDS v2024_07_08(4), and CEDS v2021_04_21(1), respectively. **(b)**, **(d)**, and **(f)** compare our emission anomalies with top-down inversions: the GCP surface-based (2017) inversions, the GCP satellite-based (2017) inversions, and the combined inversions, respectively(5).

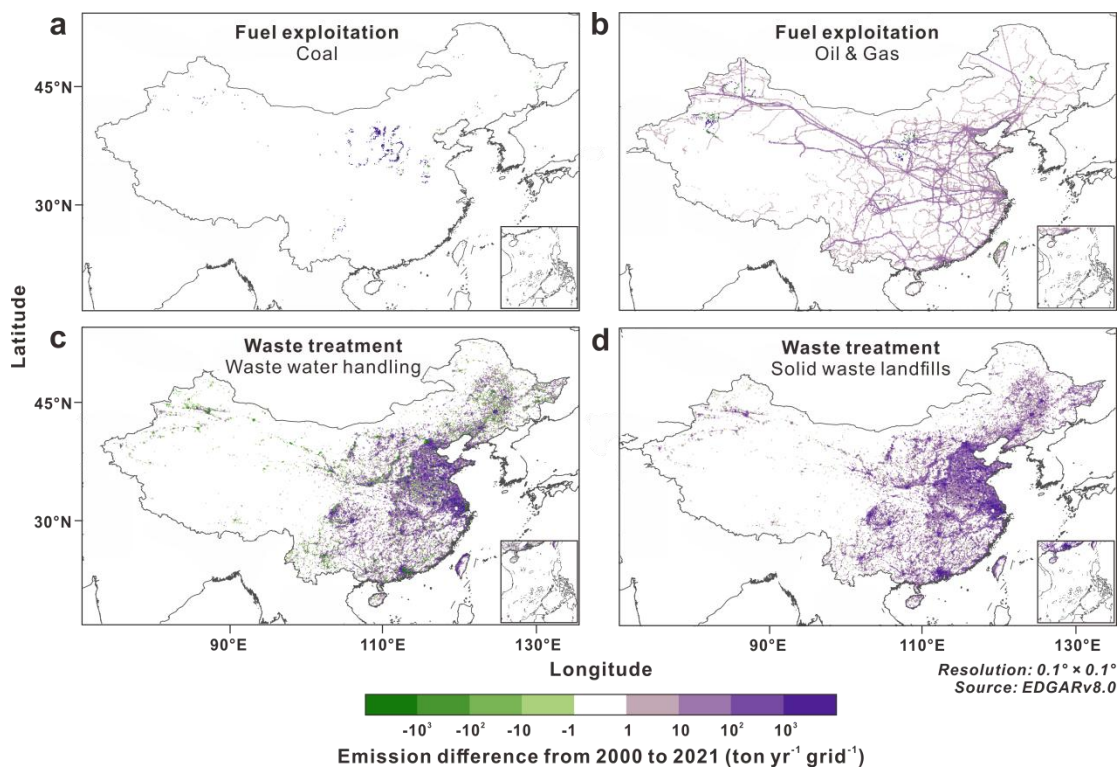


Fig. S6. Spatial distribution of CH₄ emission changes from 2000 to 2021 in key sectors of China based on the EDGARv8.0 dataset. (a–d) The spatial distribution of 21-year emission changes from the following sources: coal exploitation (a), oil and gas exploitation (b), wastewater handling (c), and solid waste landfills (d), under a spatial resolution of 0.1° × 0.1°. These four sources, within the fuel exploitation and waste treatment sectors, collectively account for over 90% of the observed emission increase during the 21 years, as derived from the EDGARv8.0 dataset (https://edgar.jrc.ec.europa.eu/dataset_ghg80).

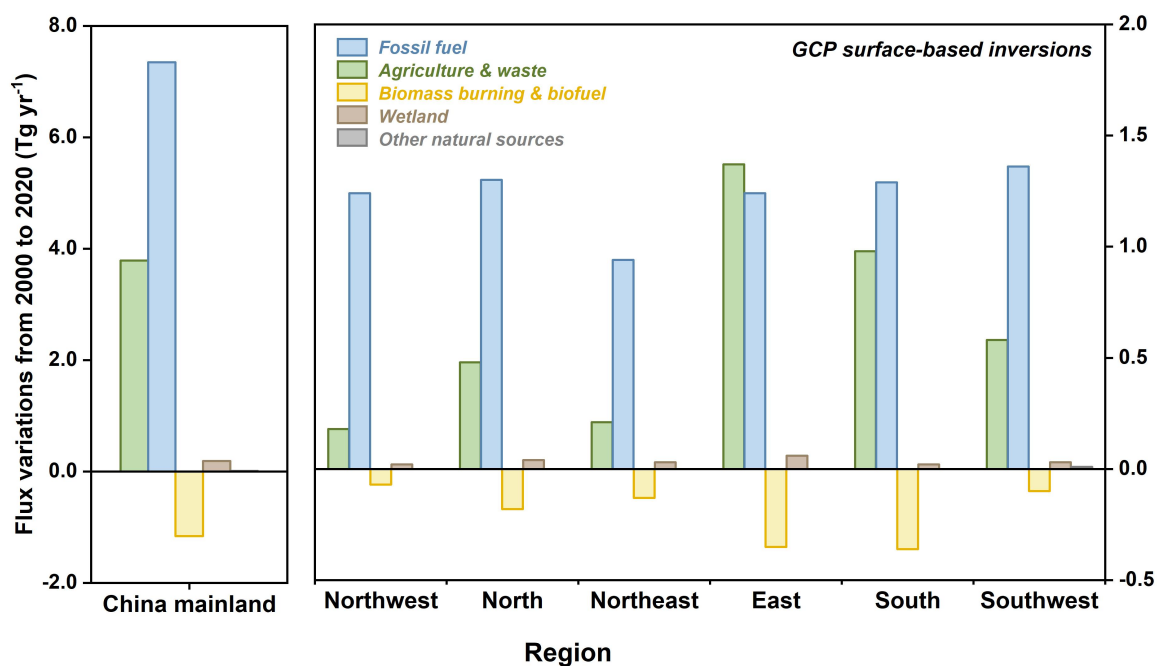


Fig. S7. Sectoral contributions to the total CH₄ flux increase in mainland China and geographical regions from 2000 to 2020 based on GCP surface-based inversions. The sectors are categorized by the GCP surface-based inversions into fossil fuels, agriculture and waste, biomass burning and biofuel, wetlands, and other natural sources. The regions analyzed include Northwest, North, Northeast, East, South, and Southwest China (Table S2). The CH₄ fluxes in China from 2000 to 2020 are extracted from the average of the GCP surface-based global CH₄ budget ensembles (6).

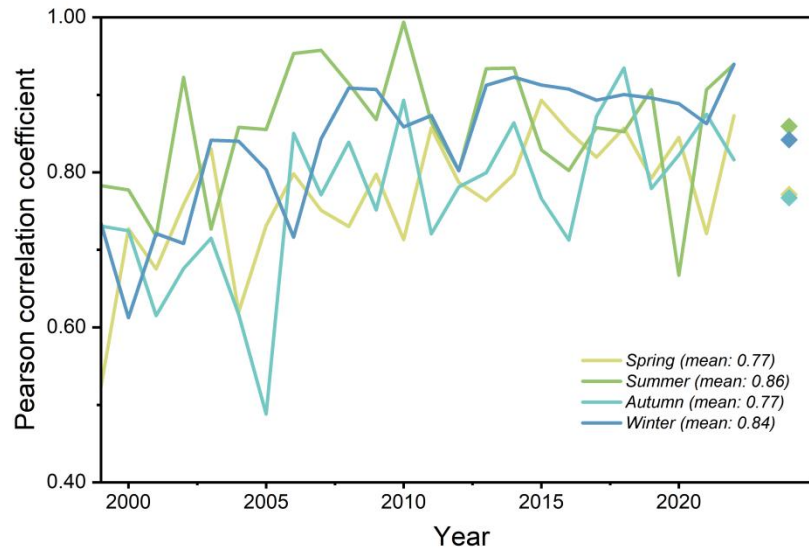


Fig. S8. Temporal trends in the Pearson correlation coefficients of each season between CH₄ and CO *EnhCs* from 1999 to 2022.

Table S1. Annual CH₄ flux estimations in China and relative estimation coefficients.

Year	t (h)	τ_{CO} (d)	τ_{CH_4} (yr)	CF	F_{CO} (Tg yr ⁻¹)	$EnhR$	F_{CH_4} (Tg yr ⁻¹)
2000	91.4	48.8	12.0	0.926	245	0.312	40.5
2001	89.4	48.4	12.0	0.927	215	0.342	38.9
2002	90.3	47.9	12.0	0.925	239	0.289	36.5
2003	87.2	47.4	12.0	0.927	243	0.297	38.2
2004	79.1	46.9	12.0	0.933	252	0.277	37.3
2005	72.9	46.5	12.0	0.937	252	0.284	38.3
2006	98.3	46.0	12.0	0.916	252	0.307	40.5
2007	90.9	45.6	12.0	0.921	252	0.348	46.2
2008	73.1	45.1	12.0	0.935	229	0.370	45.2
2009	95.3	44.7	12.0	0.916	226	0.378	44.8
2010	77.2	44.2	12.0	0.931	232	0.374	46.2
2011	88.6	43.8	12.0	0.920	237	0.418	52.2
2012	91.9	43.3	12.0	0.916	222	0.424	49.2
2013	88.7	42.9	12.0	0.918	221	0.404	46.7
2014	88.5	42.5	12.0	0.918	207	0.465	50.5
2015	92.1	42.1	12.0	0.914	192	0.494	49.5
2016	91.3	41.7	12.0	0.914	181	0.581	55.1
2017	93.6	41.3	12.0	0.911	188	0.585	57.3
2018	95.7	40.9	12.0	0.908	171	0.632	56.0
2019	84.3	40.5	12.0	0.918	171	0.683	61.2
2020	102	40.1	12.0	0.900	172	0.714	63.2
2021	86.6	39.7	12.0	0.914	170	0.754	67.1

* t is the travel time; τ_{CO} and τ_{CH_4} are lifetimes of CO and CH₄; CF is the conversion factor; F_{CO} and F_{CH_4} are the fluxes of CO and CH₄; and $EnhR$ is the enhancement ratio of CH₄ to CO.

Table S2. Regional distribution of provinces in mainland China

Region	Provinces
North China	Beijing, Tianjin, Shanxi, Hebei, Inner Mongolia
East China	Shanghai, Jiangsu, Zhejiang, Anhui, Fujian, Jiangxi, Shandong, Taiwan
Northwest China	Shannxi, Gansu, Qinghai, Ningxia, Xinjiang
Southwest China	Sichuan, Guizhou, Yunnan, Chongqing, Tibet
South China	Guangdong, Guangxi, Hainan, Henan, Hubei, Hunan
Northeast China	Heilongjiang, Jilin, Liaoning

Source: (7)

Table S3. The detailed settings of the sensitivity test

Setting contents	Setting objective
<i>Enhancement Ratio (EnhR)</i>	
NCEP/NCAR global reanalysis data (Reference inversion)	The impact of different meteorological fields in the HYSPLIT model on the CH ₄ flux estimation
The Global Data Assimilation System (GDAS) (Meteorology_GDAS)	
Height: 500 m (Reference inversion)	The impact of different height settings in the HYSPLIT model on the CH ₄ flux estimation
Height: 100 m (Height_100 m)	
Height: 1000 m (Height_1000 m)	
Height: 1500 m (Height_1500 m)	
Height: 2000 m (Height_2000 m)	
Perform a linear fit on all values and apply the three-year moving average method (Reference inversion)	The impact of different data processing of linear fitting on the CH ₄ flux estimation
Perform a linear fit on values within two standard errors and apply the three-year moving average method (Exclude data beyond 2std)	
Perform a linear fit on all values weighted by total CH ₄ emissions along each trajectory and apply the three-year moving average method (Weighted_emissions)	
Perform a linear fit on all values without the three-year moving average method (No moving average)	
<i>Conversion Factor (CF)</i>	
Obtaining interannual variations in travel time based on different weather conditions and emission scenarios (Reference inversion)	The impact of travel time on the CH ₄ flux estimation
Fixed travel time (Fixed travel time)	
CO and CH ₄ lifetimes under varying temperature and OH concentrations (Reference inversion)	The impact of gases' lifetimes on the CH ₄ flux estimation
Fixed CO lifetime and varying CH ₄ lifetimes (Fixed CO lifetime)	
Fixed CH ₄ lifetime and varying CO lifetimes (Fixed CH ₄ lifetime)	
CO and CH ₄ lifetimes under varying OH concentrations and fixed atmospheric temperature (Fixed temperature)	
CO and CH ₄ lifetimes under fixed OH concentration and varying atmospheric temperature (Fixed [OH])	

SI References

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2. B. Zheng *et al.*, Global atmospheric carbon monoxide budget 2000–2017 inferred from multi-species atmospheric inversions. *Earth Syst. Sci. Data* **11**, 1411-1436 (2019).
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6. M. Saunio *et al.*, Global Methane Budget 2000-2020. *Earth Syst. Sci. Data Discuss.* **2024**, 1-147 (2024).
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