

1 *Supplementary Materials for*

2 **Signal-Domain Guided Deep Learning for Gap-Filling of**
3 **XCO and XCH₄: A Masked Spatio-Temporal Fusion of**
4 **TROPOMI and GEOS-Chem (2019–2023)**

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Supplementary tables

Table S1. Site-based validation results of XCO using different reconstruction methods. Metrics include coefficient of determination (R^2), root mean square error (RMSE, ppb), and mean bias (μ , ppb).

Column	R^2	RMSE	μ
Only DCT	0.8227	10.439	-3.815
Only Mixed-signal	0.8335	10.1166	-3.7682
Residual CNN	0.8385	9.9619	-3.7677
Residual U-net	0.845	9.7616	-3.686
Residual XGBoost	0.8392	9.9406	-3.7435

Table S2. Site-based validation results of XCH₄ using different reconstruction methods. Metrics include coefficient of determination (R^2), root mean square error (RMSE, ppb), and mean bias (μ , ppb).

Column	R^2	RMSE	μ
Only DCT	0.7056	17.5156	-6.9345
Only Mixed-signal	0.732	16.7111	-6.9635
Residual CNN	0.7469	16.2404	-6.9162
Residual U-net	0.7598	15.8212	-6.7858
Residual XGBoost	0.7476	16.2188	-6.9563

Table S3. (GEOS) Single-site validation results for GEOS-Chem, TROPOMI, and fused XCO data, with RMSE & μ in parts per billion (ppb).

SITE NAME	R^2			RMSE			μ		
	GEOS	TROPOMI	FUSION	GEOS	TROPOMI	FUSION	GEOS	TROPOMI	FUSION
burgos01	0.237	0.919	0.935	16.909	5.769	5.246	9.667	-2.799	-2.425
darwin01	-0.808	0.370	0.646	13.896	8.546	6.330	-9.241	-4.196	-2.848
easttroutlake01	-0.487	0.827	0.903	19.017	10.070	7.372	4.540	0.688	1.092
garmisch01	0.664	0.795	0.806	5.903	3.897	4.444	-1.453	0.737	1.113
hefei01	0.583	0.930	0.980	16.501	7.362	3.421	-15.897	-1.450	1.444
izana01	0.093	0.185	0.513	6.943	7.120	5.482	-3.692	5.790	3.909
karlsruhe01	0.521	0.411	0.596	7.055	6.892	6.441	4.002	-3.137	-4.337
lamont01	-0.134	0.534	0.580	9.297	6.447	6.013	5.209	-4.803	-3.650
lauder03	-0.524	0.849	0.663	11.990	3.893	5.666	-8.828	-2.228	-2.424
nicosia01	-0.251	0.868	0.847	8.114	2.791	3.008	5.978	0.557	-1.147
orleans01	0.581	0.791	0.537	6.704	6.656	8.947	3.012	-2.665	-2.786
parkfalls01	-0.468	0.899	0.811	19.445	6.597	8.677	7.525	-2.116	-2.106
reunion01	0.109	0.893	0.896	8.529	3.113	3.061	2.327	0.448	-0.859
rikubetsu01	0.635	0.961	0.899	7.674	2.893	4.545	1.122	0.064	0.438
sodankyla01	0.388	0.267	0.917	6.255	6.203	2.230	-0.775	2.807	1.381
tsukuba02	0.955	0.825	0.958	4.446	9.713	4.031	-2.817	-6.798	-1.684
wollongong01	-0.319	0.776	0.745	14.709	6.562	6.969	-5.625	-4.501	-4.418
xianghe01	0.279	0.269	0.553	11.965	10.749	9.469	-2.922	6.306	2.628

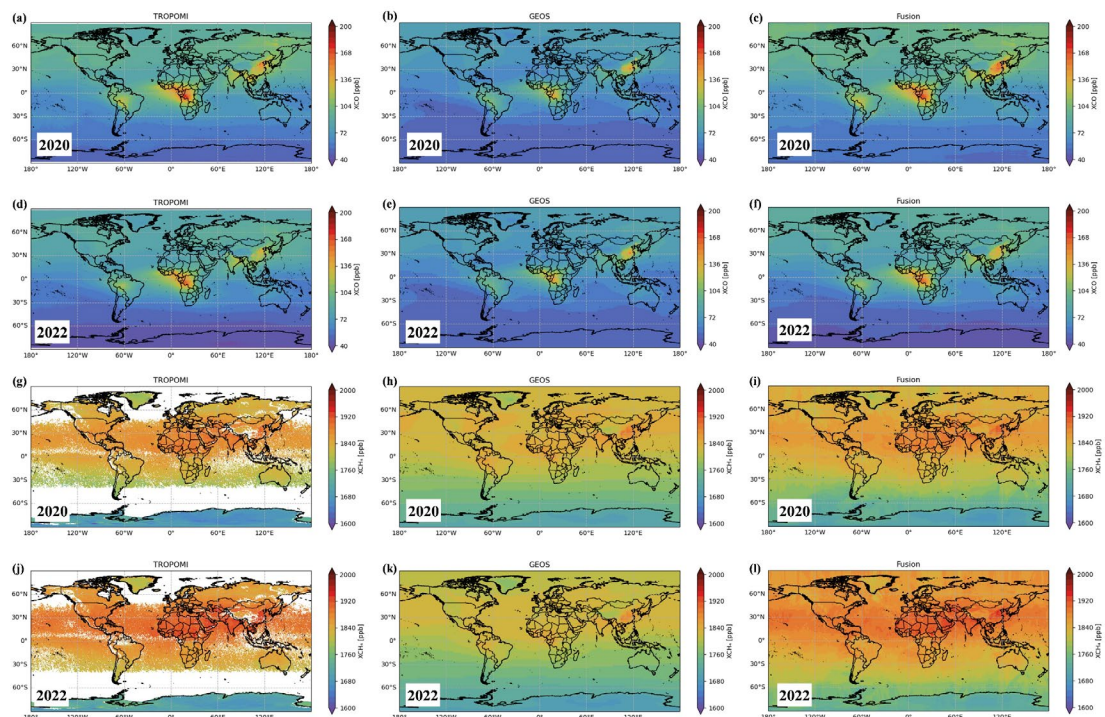
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Table S4. (GEOS) Single-site validation results for GEOS-Chem, TROPOMI, and fused XCH₄ data, with RMSE & μ in parts per billion (ppb).

SITE NAME	R ²			RMSE			μ		
	GEOS	TROPOMI	FUSION	GEOS E	TROPOMI	FUSION	GEOS	TROPOMI	FUSION
bremen01	-0.140	0.134	0.631	13.602	10.634	8.139	-0.024	-6.005	-2.375
burgos01	-0.280	0.741	0.877	24.716	13.217	8.892	-11.397	-7.712	-5.010
easttroutlake01	-0.560	0.371	0.322	27.858	18.152	20.471	-7.800	-10.457	-12.565
edwards01	-1.666	0.938	0.908	33.363	5.655	6.750	-20.863	2.314	4.679
garmisch01	-0.539	0.693	0.831	23.574	11.552	8.763	-4.177	-4.162	-3.266
hefei01	-1.130	0.882	0.770	38.129	8.789	12.575	-26.749	-3.298	-5.167
karlsruhe01	-1.466	0.546	0.561	31.590	13.937	14.156	-17.057	-8.526	-11.191
lamont01	-2.086	0.838	0.830	34.635	9.308	9.313	-22.403	-3.808	-4.785
lauder03	-1.178	0.725	0.742	33.162	12.366	12.116	-16.148	-7.392	-9.347
nicosia01	-2.566	0.670	0.893	32.223	10.568	5.954	-23.891	2.413	-1.740
orleans01	-1.780	0.551	0.705	30.640	13.366	11.070	-16.560	-6.472	-6.654
parkfalls01	-0.923	0.137	0.221	29.718	21.329	20.668	-12.079	-14.704	-16.099
pasadena01	-3.177	0.955	0.910	41.561	4.659	6.603	-32.970	-0.942	-3.476
rikubetsu01	0.230	0.740	0.715	15.539	9.189	9.892	6.588	-2.146	-4.784
tsukuba02	-0.582	0.777	0.712	15.420	6.238	7.147	-1.677	-2.308	-3.793
wollongong01	-1.396	0.195	0.347	29.981	19.964	17.320	-14.676	-12.975	-13.672
xianghe01	0.033	0.855	0.888	25.528	10.349	9.120	-9.444	-0.354	-1.247

34 **Supplementary figures**



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36 **Figure S1.** (a, d) Annual global average concentration distribution of XCO data from TROPOMI,
37 (b, e) GEOS-Chem, and (c, f) fused data; (g, i) Annual global average concentration distribution of
38 XCH₄ data from TROPOMI, (h, k) GEOS-Chem, and (i, l) fused data. The color bar represents the
39 concentrations of XCO and XCH₄, with units in parts per billion (ppb).

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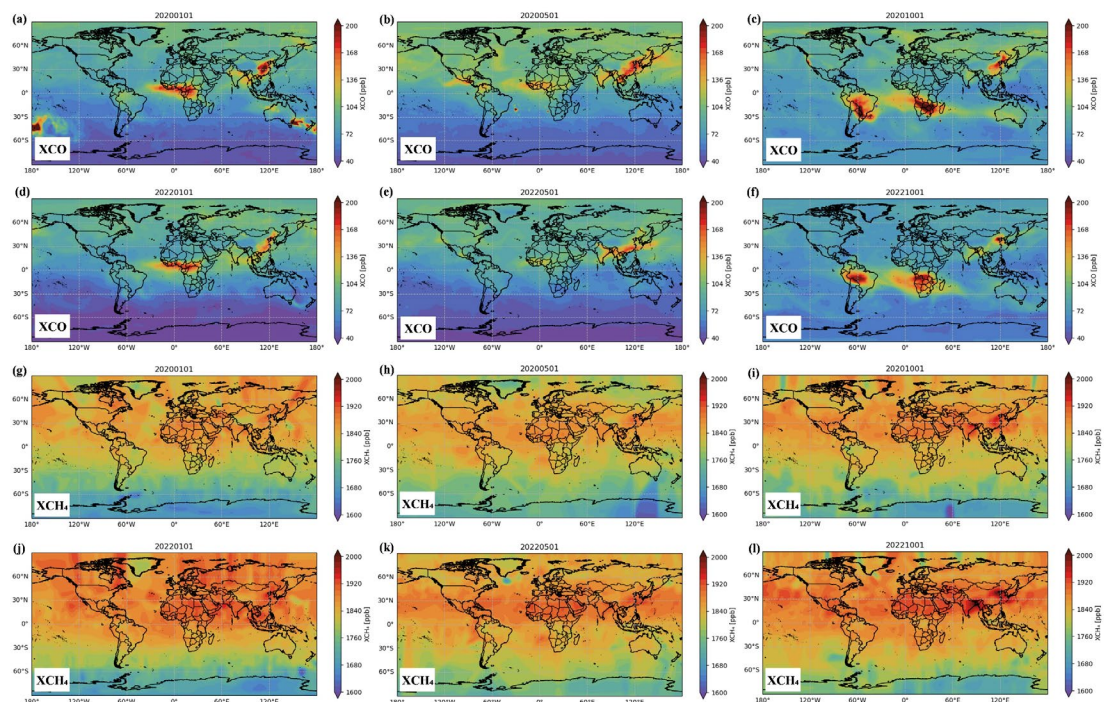


Figure S2. (a-f) Single-day global concentration distribution of the fused XCO data; (g-l) Single-day global mean concentration distribution of the fused XCH₄ data. The color bars indicate the concentrations of XCO and XCH₄ in parts per billion (ppb).

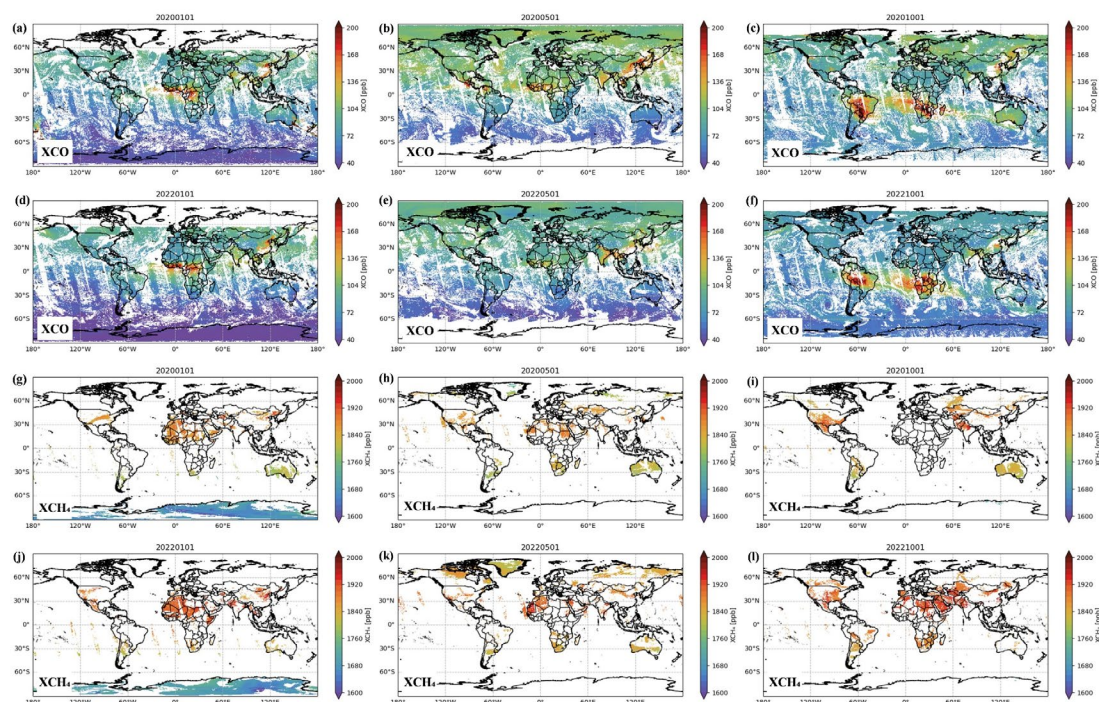


Figure S3. (a-f) Distribution of single-day global concentrations of XCO observed by TROPOMI; (g-l) Distribution of single-day global mean concentrations of XCH₄ observed by TROPOMI. The color bars indicate the concentrations of XCO and XCH₄ in parts per billion (ppb).

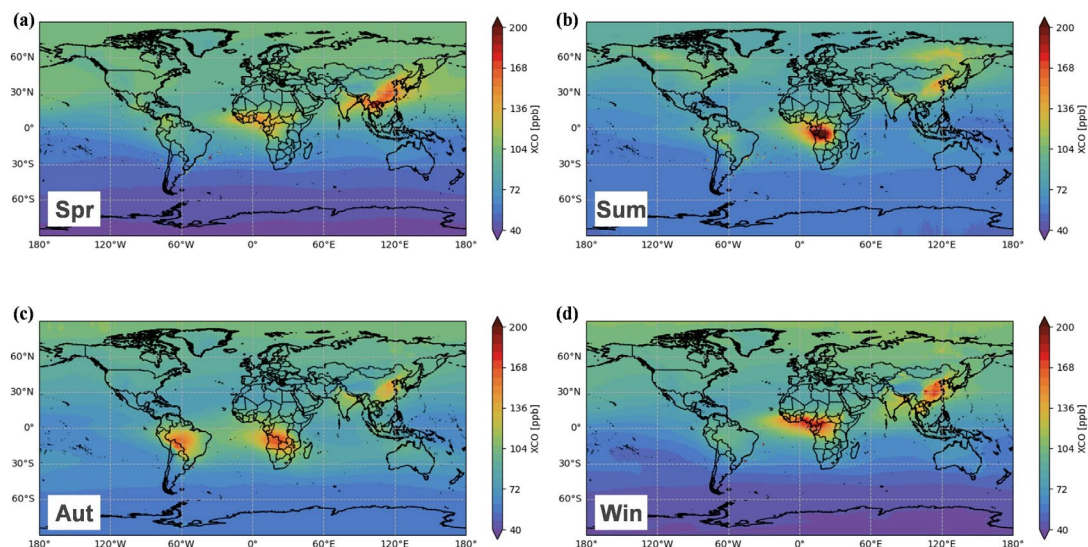


Figure S4. Seasonal fusion data of XCO from 2019 to 2023 in global concentration distribution, color bars indicate XCO concentrations, all in parts per billion (ppb). (a) Spr, (b) Sum, (c) Aut and (d) Win indicate December through February, March through May, June through August, and September through November, respectively.

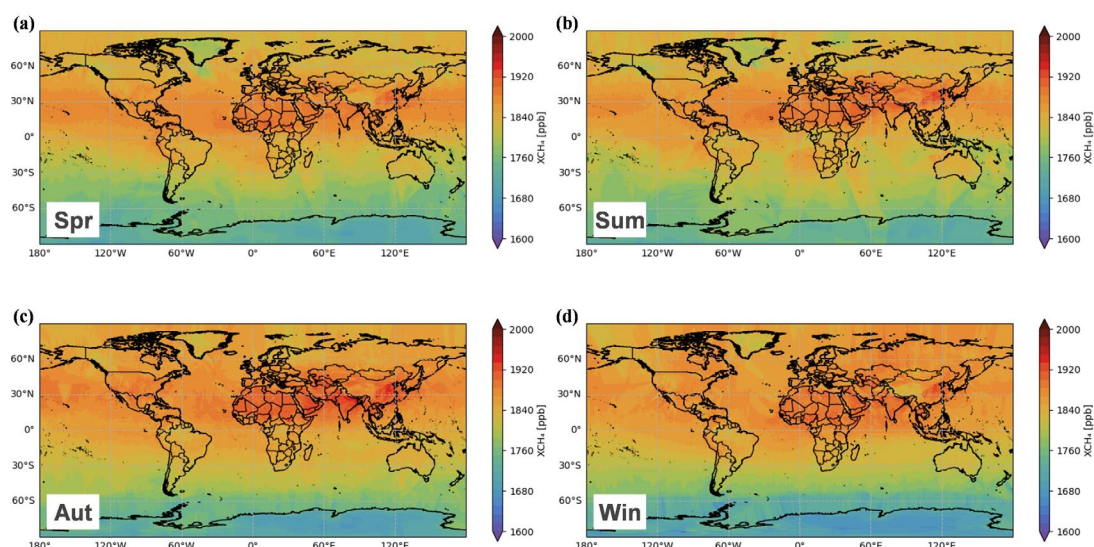


Figure S5. Seasonal fusion data of XCH₄ from 2019 to 2023 in global concentration distribution, color bars indicate XCH₄ concentrations, all in parts per billion (ppb). (a) Spr, (b) Sum, (c) Aut and (d) Win indicate December through February, March through May, June through August, and September through November, respectively.

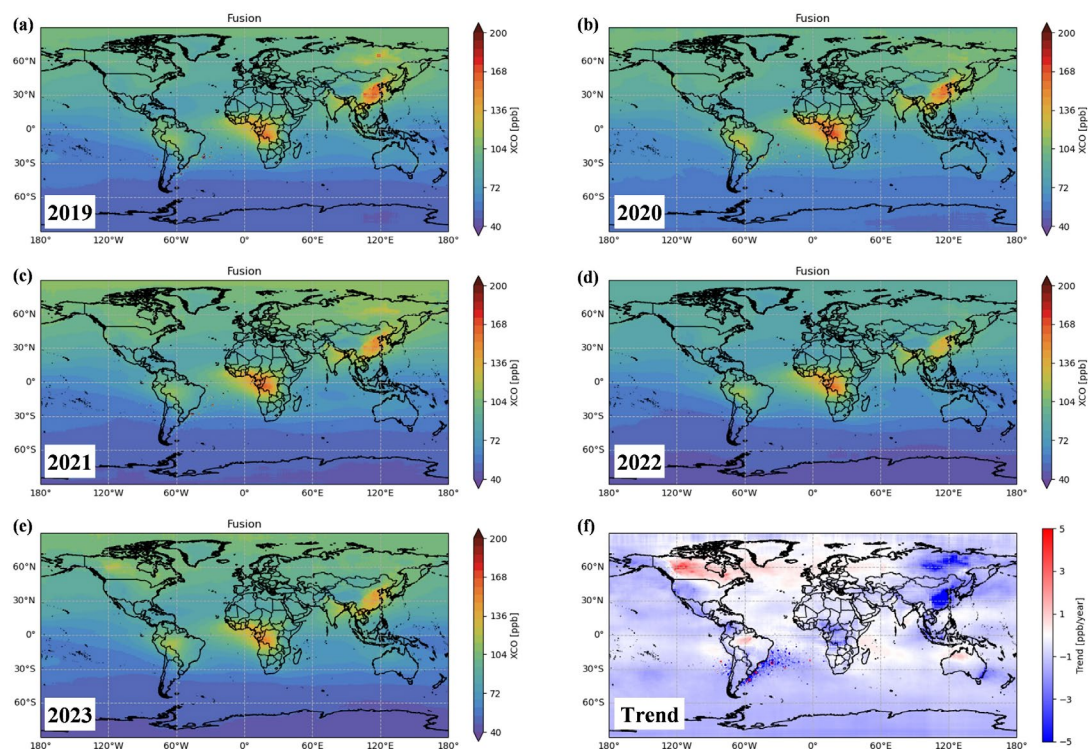


Figure S6. Global annual fusion (a-e) XCO data from 2019 to 2023, color bars indicate XCO concentration values, all in parts per billion (ppb). (f) Annual trends, color bars indicate annual trend values of XCO in parts per billion (ppb/year).

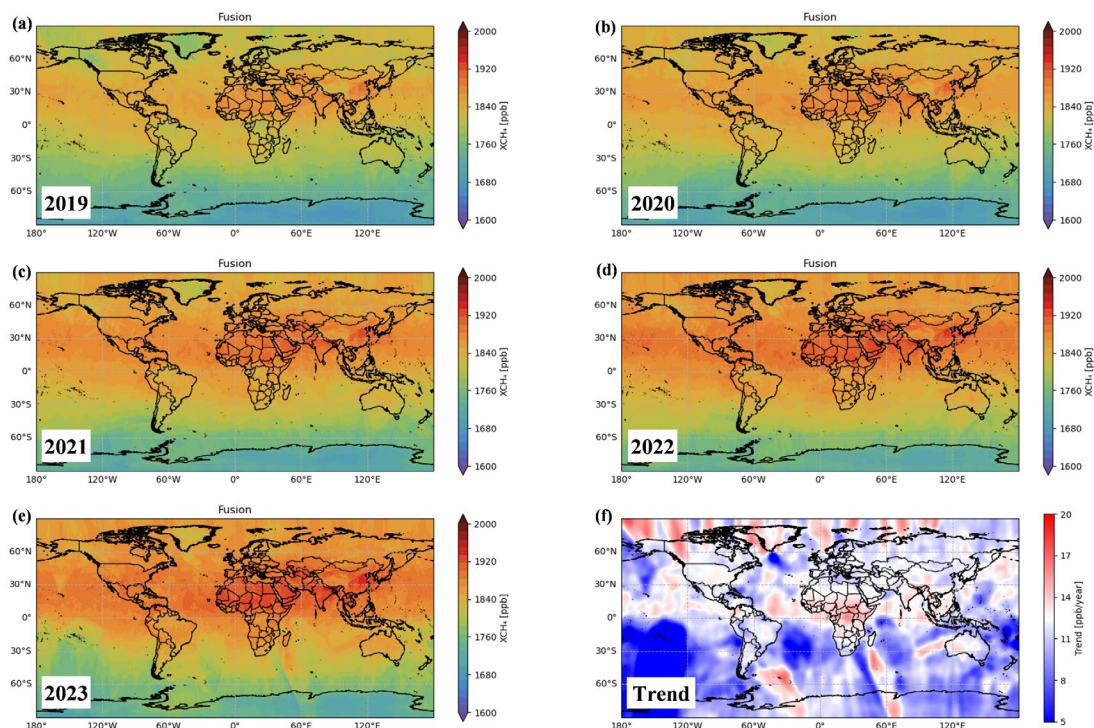


Figure S7. Global annual fusion (a-e) XCH₄ data from 2019 to 2023, color bars indicate XCH₄ concentration values, all in parts per billion (ppb). (f) Annual trends, color bars indicate annual trend values for XCH₄ in parts per billion (ppb/year).

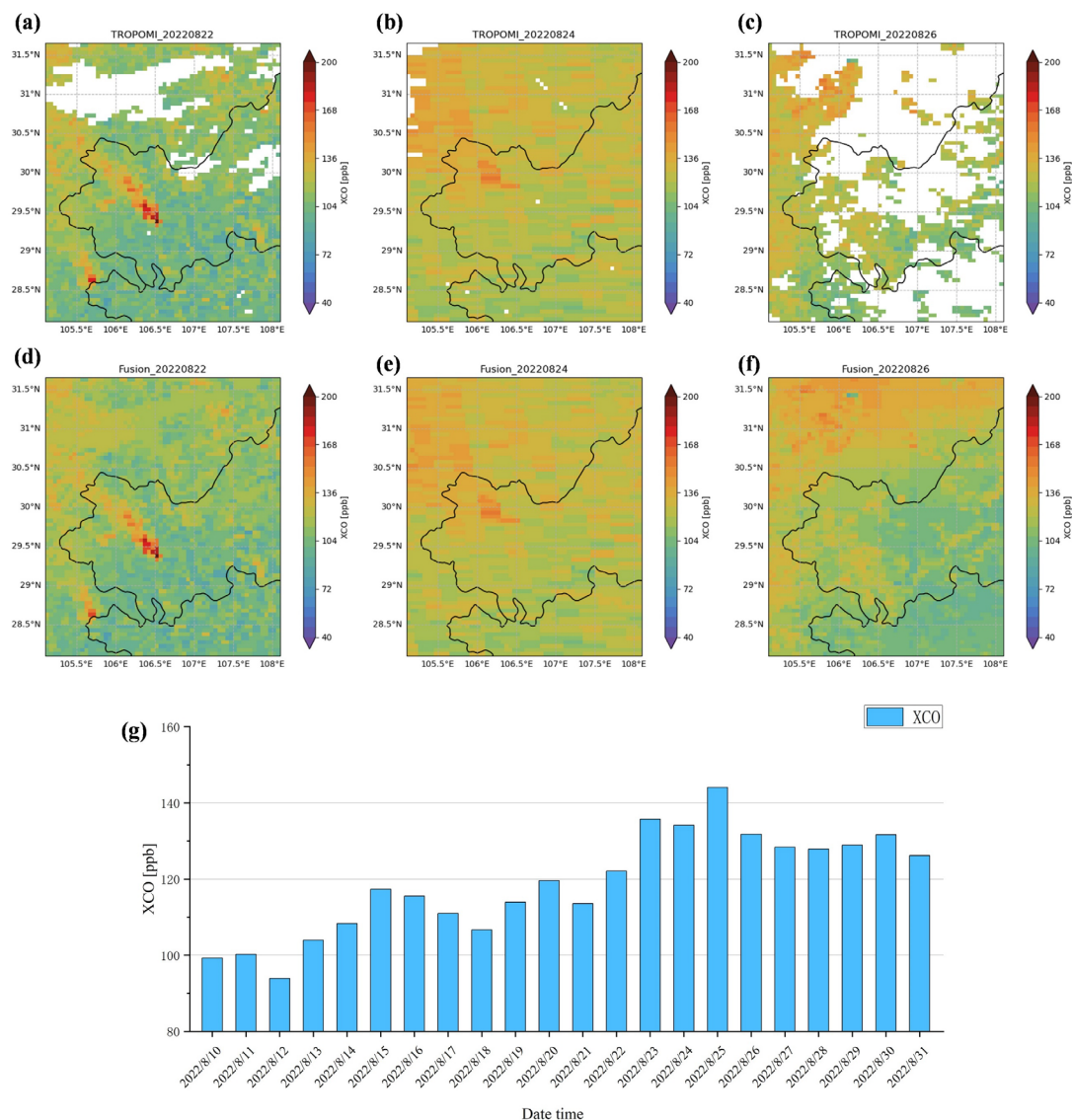


Figure S8. (a-c) TROPOMI, (d-f) Distribution of Chongqing regional concentrations on a single day of fused XCO data, color bars indicate XCO concentration values, all in parts per billion (ppb). (g) Histogram of daily concentration changes in mid- to late-August 2022 from fused XCO data, in parts per billion (ppb).

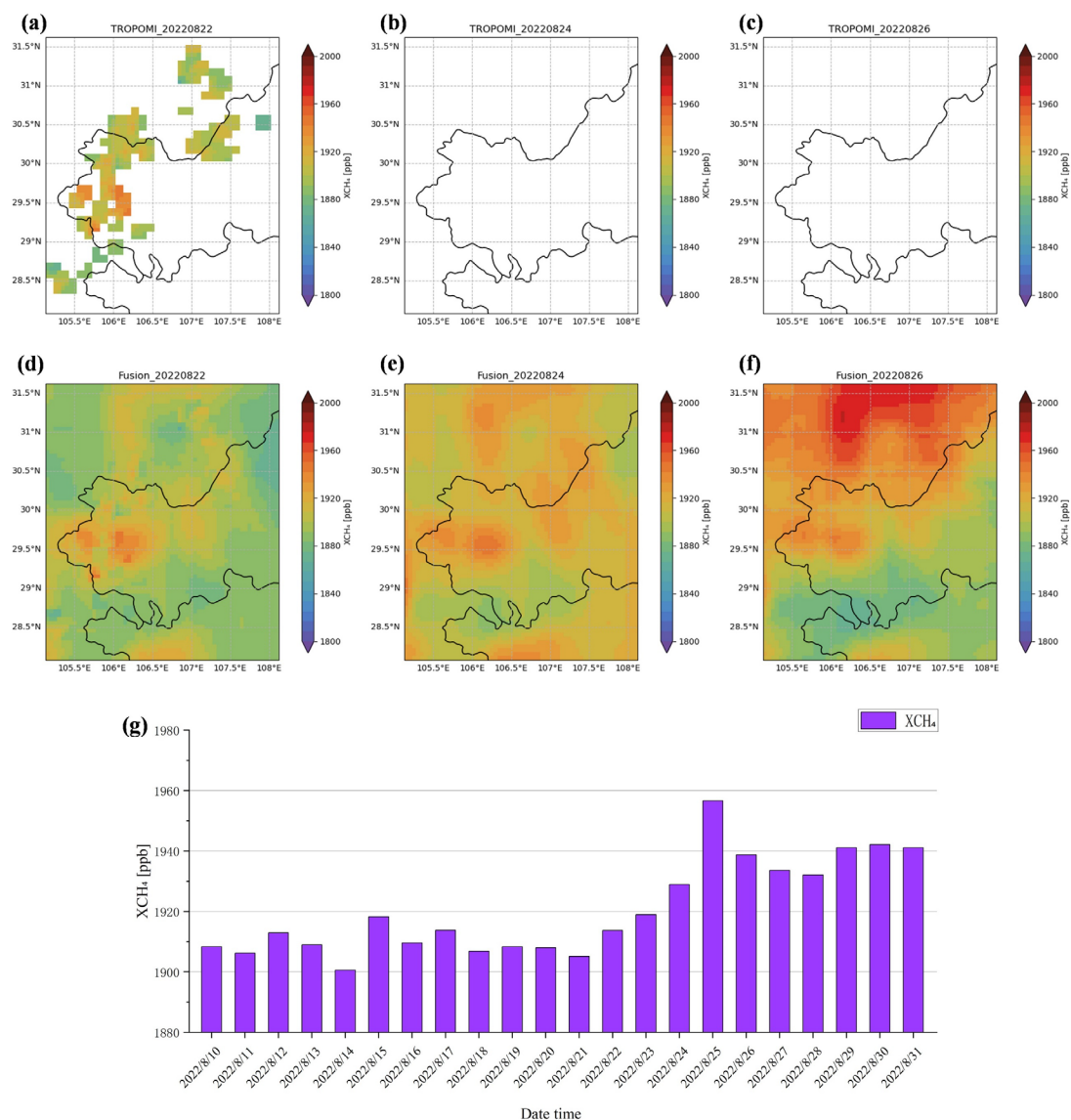


Figure S9. (a-c) TROPOMI, (d-f) Distribution of single-day Chongqing regional concentrations of fused XCH₄ data, color bars indicate XCH₄ concentration values, all in parts per billion (ppb). (g) Histogram of daily concentration changes in mid- to late-August 2022 from fused XCH₄ data, in parts per billion (ppb).