

Supplementary material for “A daily 0.025° gridded column-averaged carbon dioxide dataset for terrestrial Europe from 2019 to 2023”

Jiyuan Yang^{1,2}, Min Shao³, Jiani Tan⁴, Ruixun Xia^{1,2}, Xiangyun Gu¹, Yang Liu⁵, Andrew P. Morse², Qing Mu^{1*}

¹ Department of Health and Environmental Sciences, School of Science, Xi'an Jiaotong-Liverpool University, Suzhou 215123, China

² School of Environmental Sciences, University of Liverpool, Liverpool L69 7ZT, UK

³ School of Environment, Nanjing Normal University, Nanjing 210098, China

⁴ School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China

⁵ School of AI and Advanced Computing, Xi'an Jiaotong-Liverpool University, Suzhou 215400, China

Correspondence to: Qing Mu (qing.mu@xjtlu.edu.cn)

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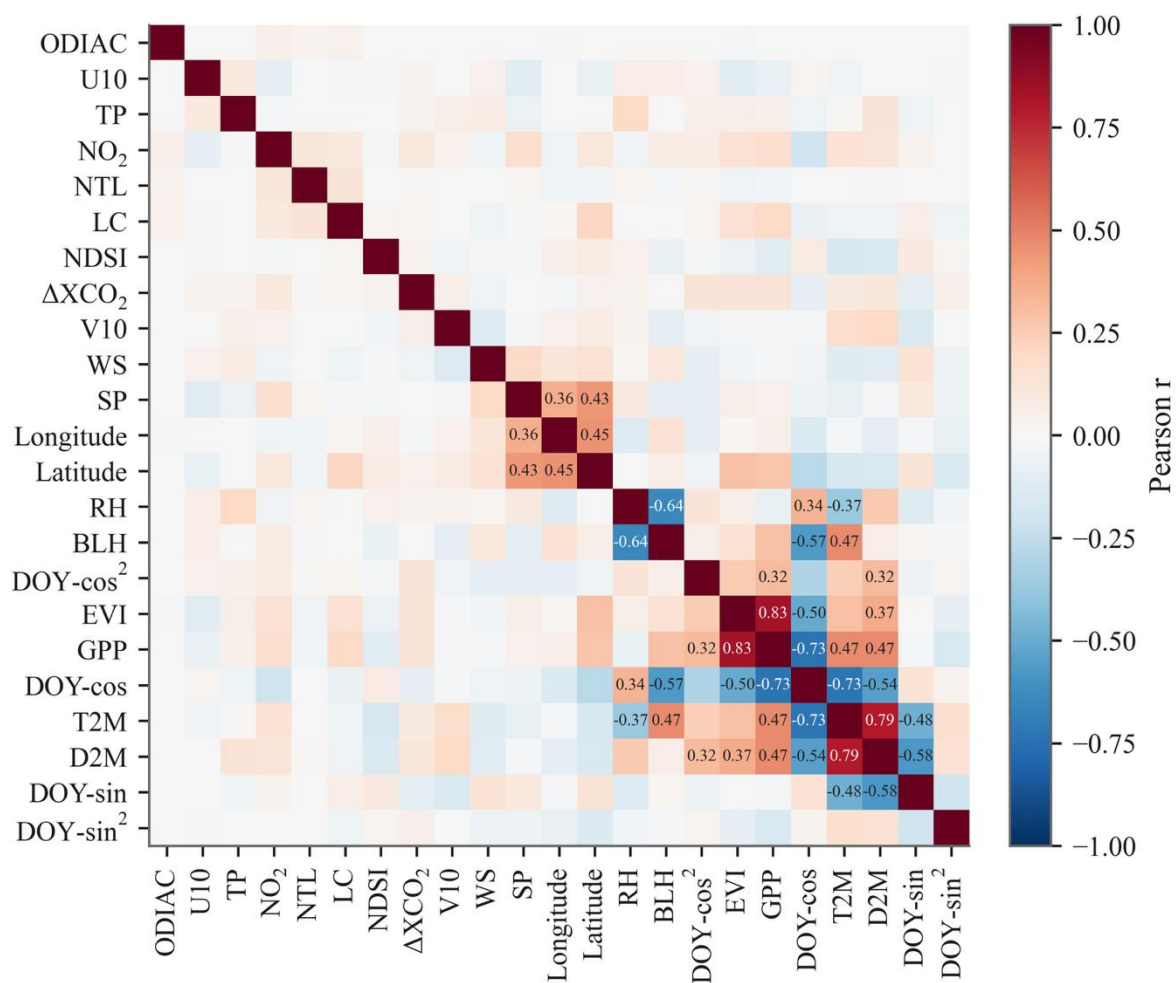


Figure S1. Pearson correlation coefficient matrix of the candidate input variables.

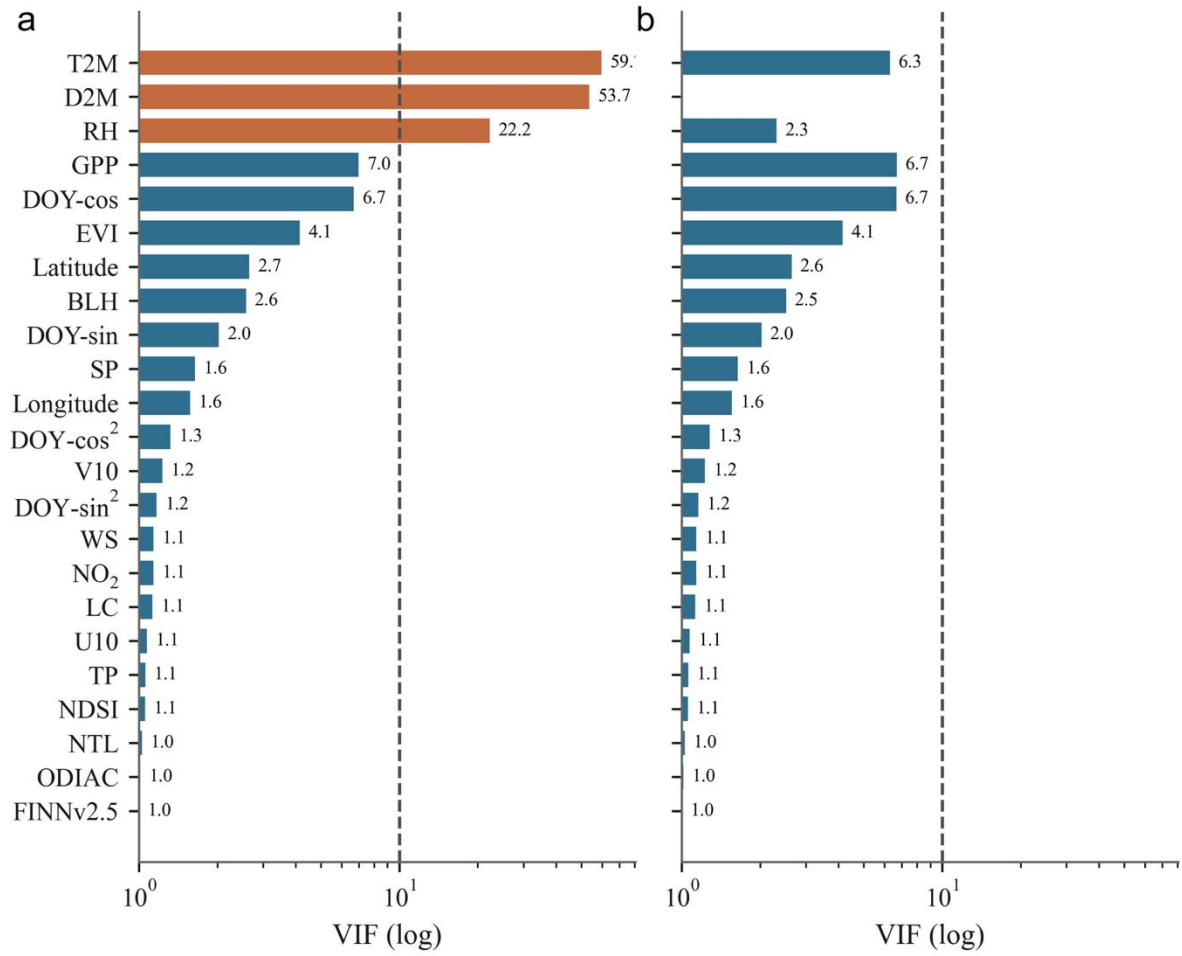


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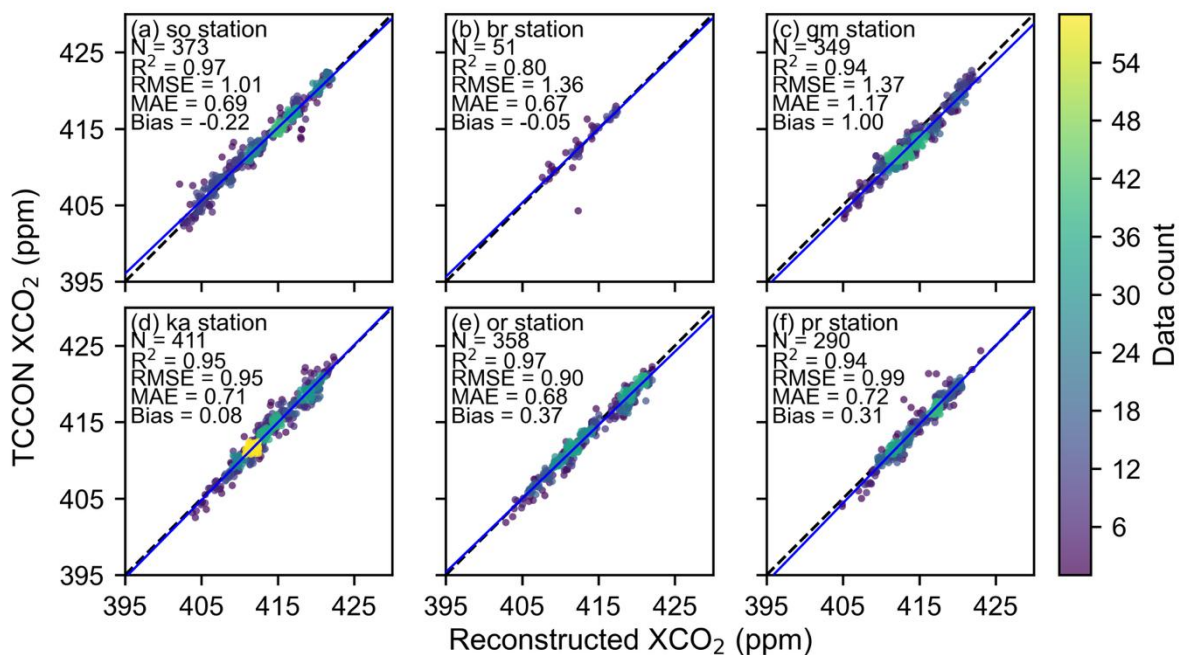


Figure S3. Comparison of our reconstructed XCO₂ dataset with each TCCON station observation^{1,2}. The black dashed line indicates the 1:1 reference, and the blue solid line shows the best-fit linear regression. The data volume is represented by color shading (see color bar for count values).

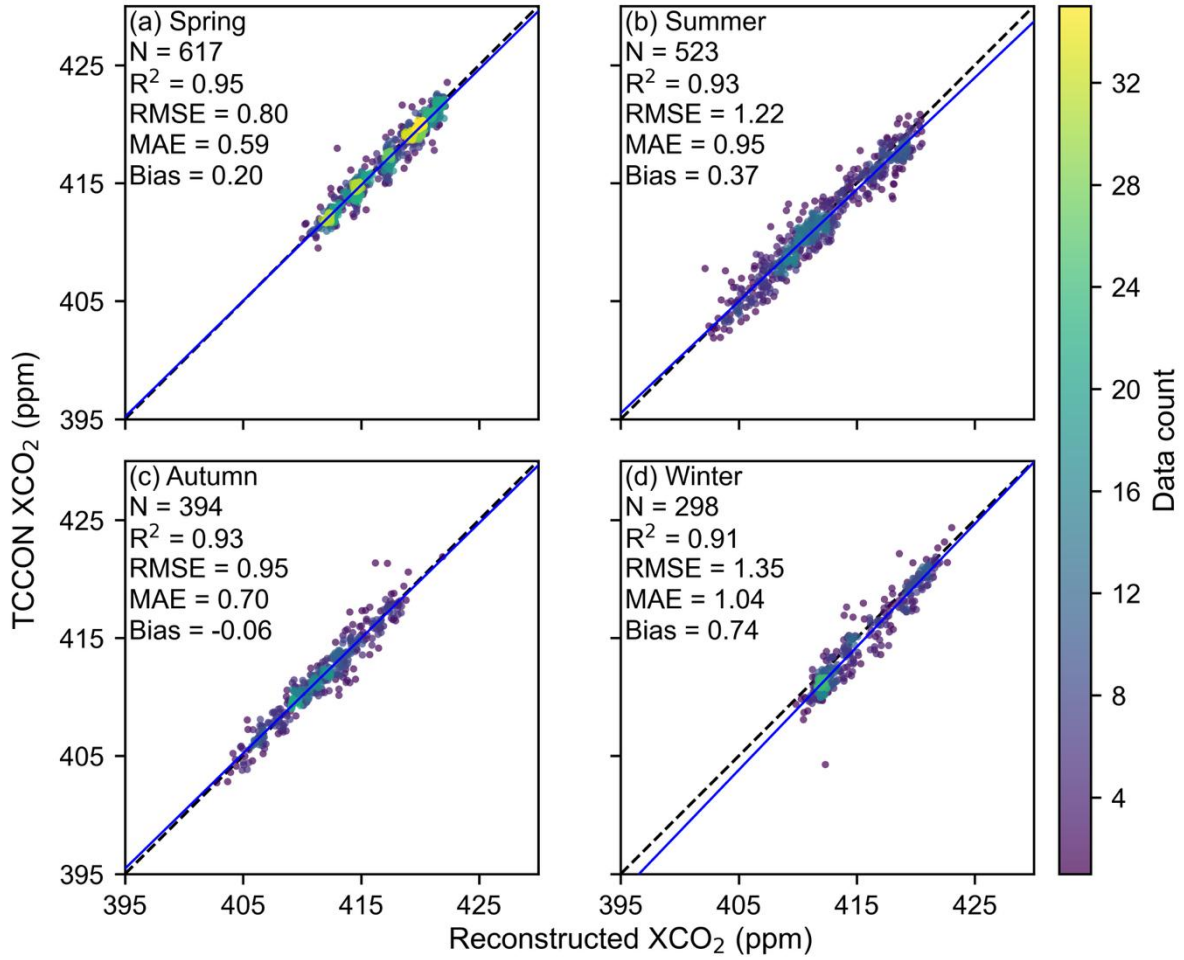
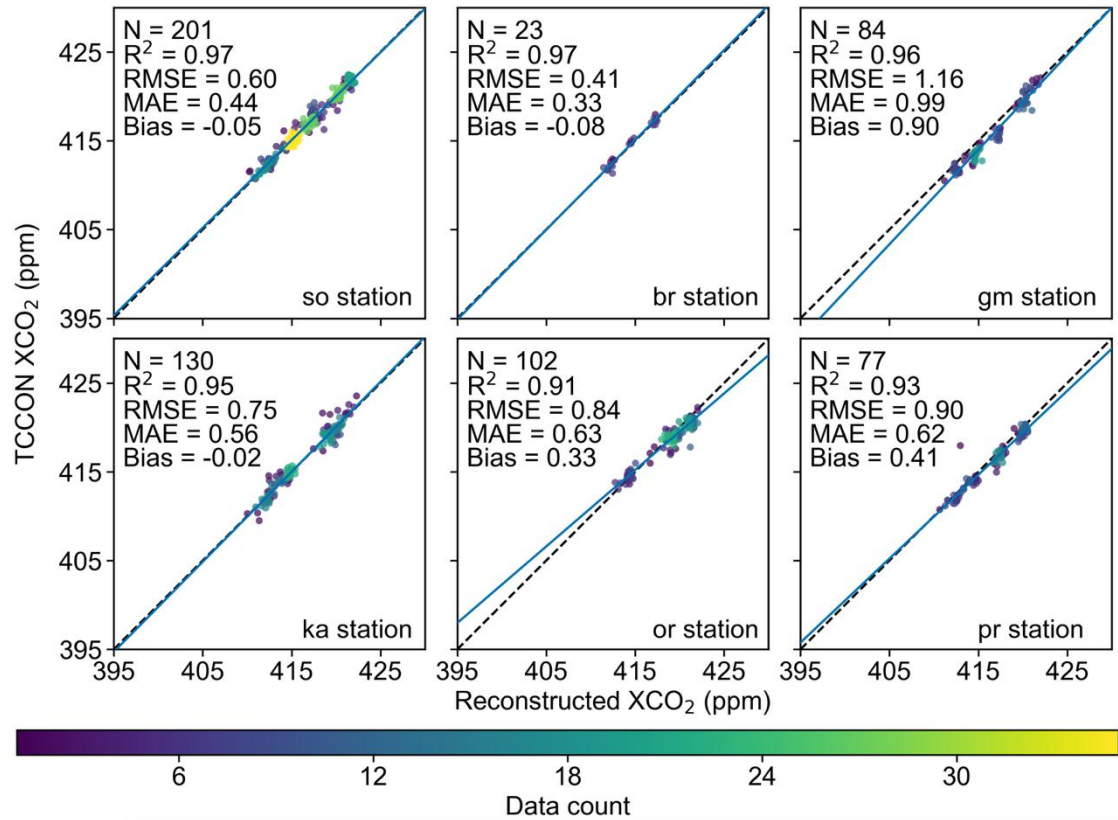
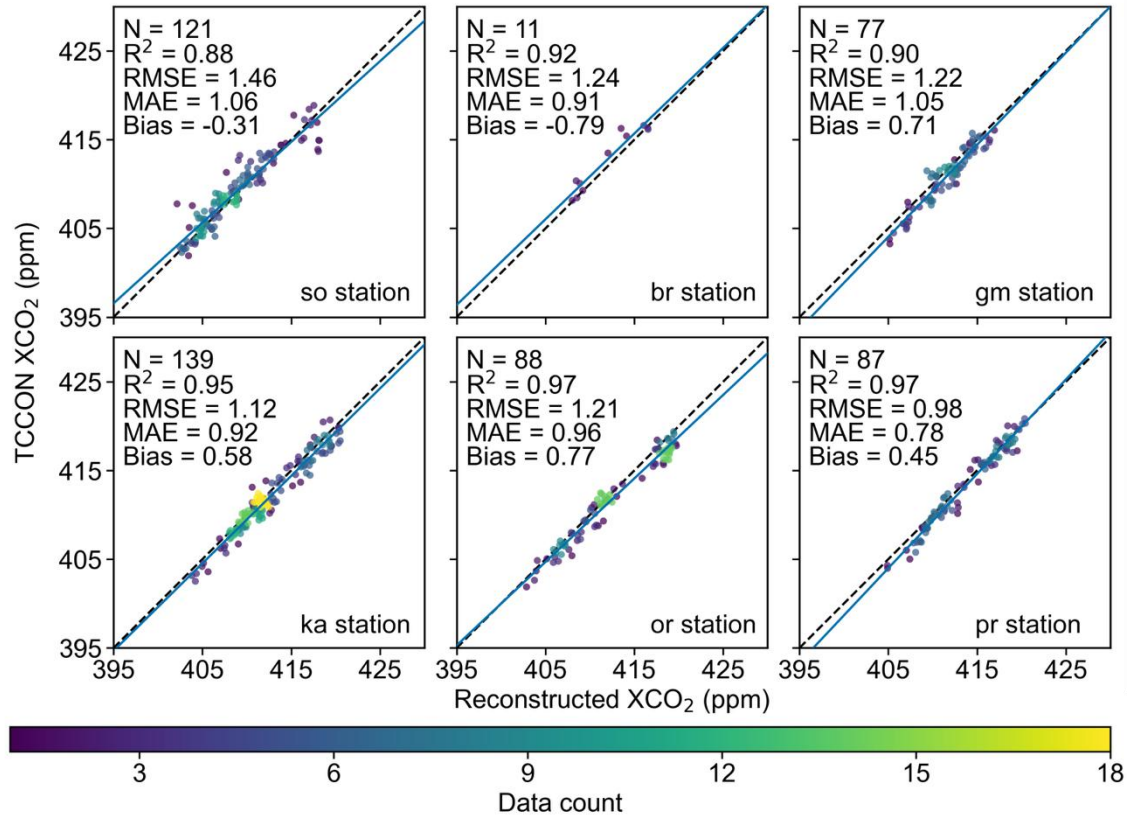


Figure S4. Comparison of our reconstructed XCO₂ dataset with TCCON observations per season. The black dashed line indicates the 1:1 reference, and the blue solid line shows the best-fit linear regression. The data volume is represented by color shading (see color bar for count values).

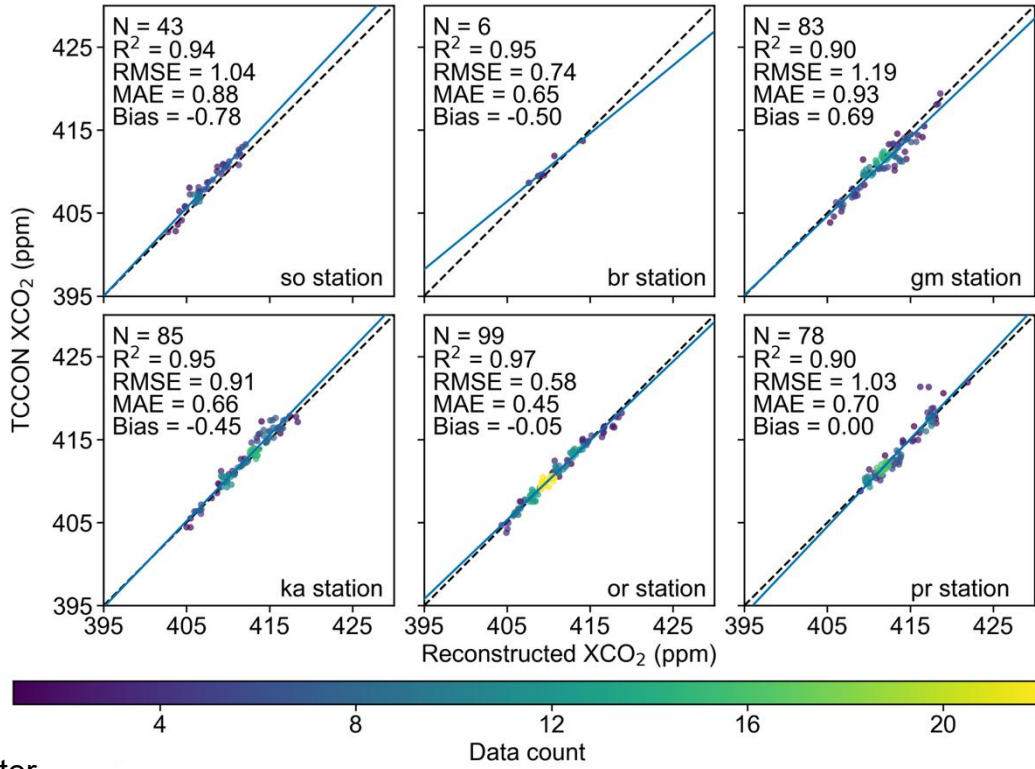
(a) Spring



(b) Summer



(c) Autumn



(d) Winter

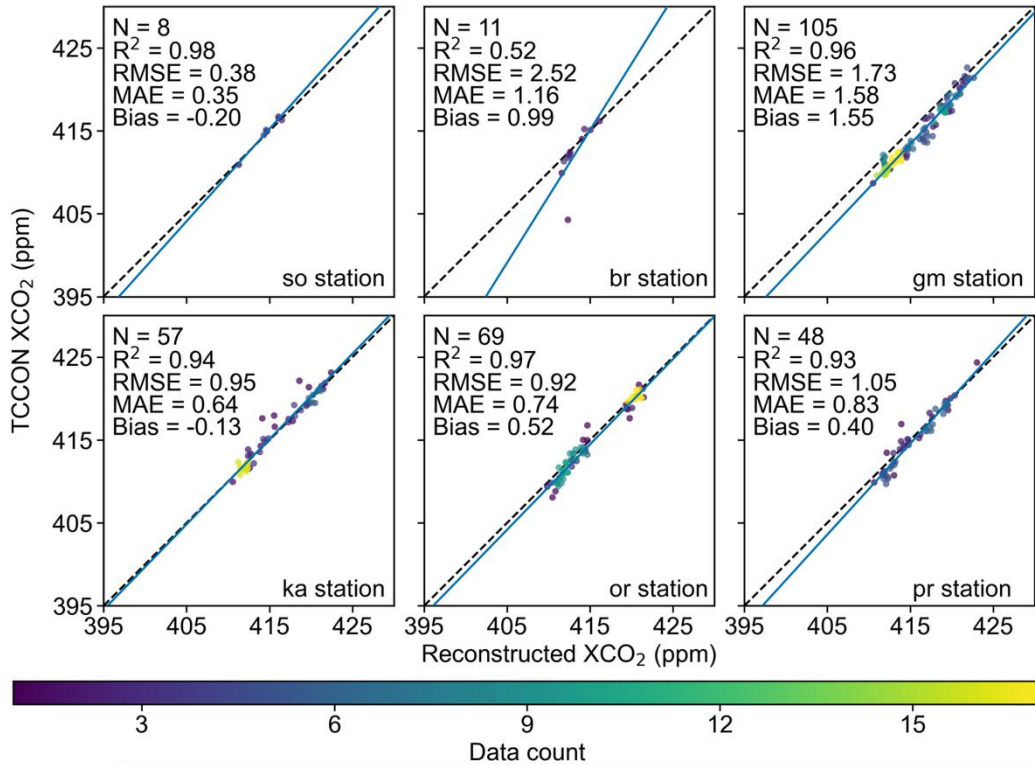


Figure S5. Comparison of our reconstructed XCO_2 dataset with TCCON observations at each station per season. (a) Spring, (b) Summer, (c) Autumn, (d) Winter. The black dashed line indicates the 1:1 reference, and the blue solid line shows the best-fit linear

regression. The data volume is represented by color shading (see color bar for count values).

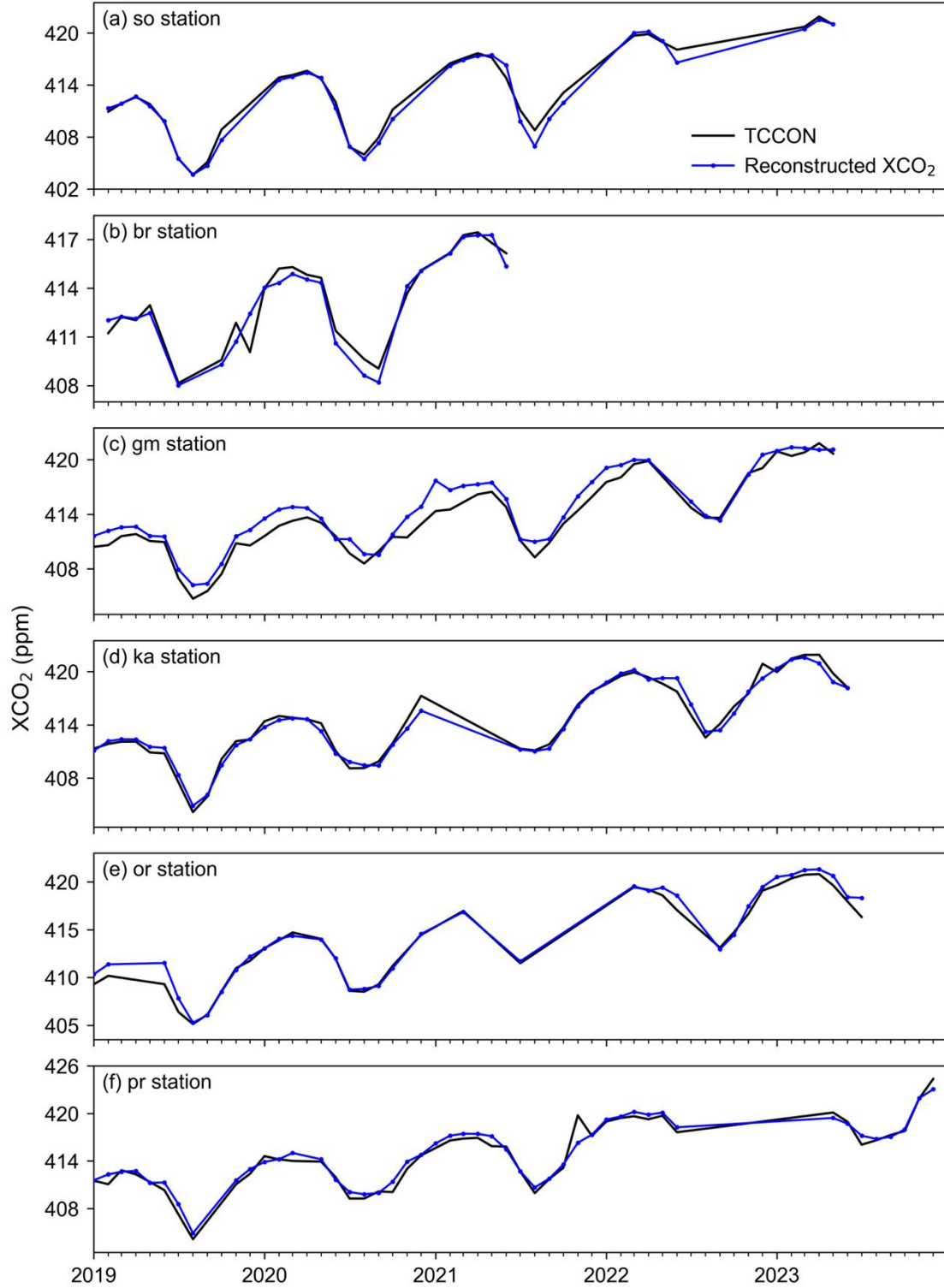


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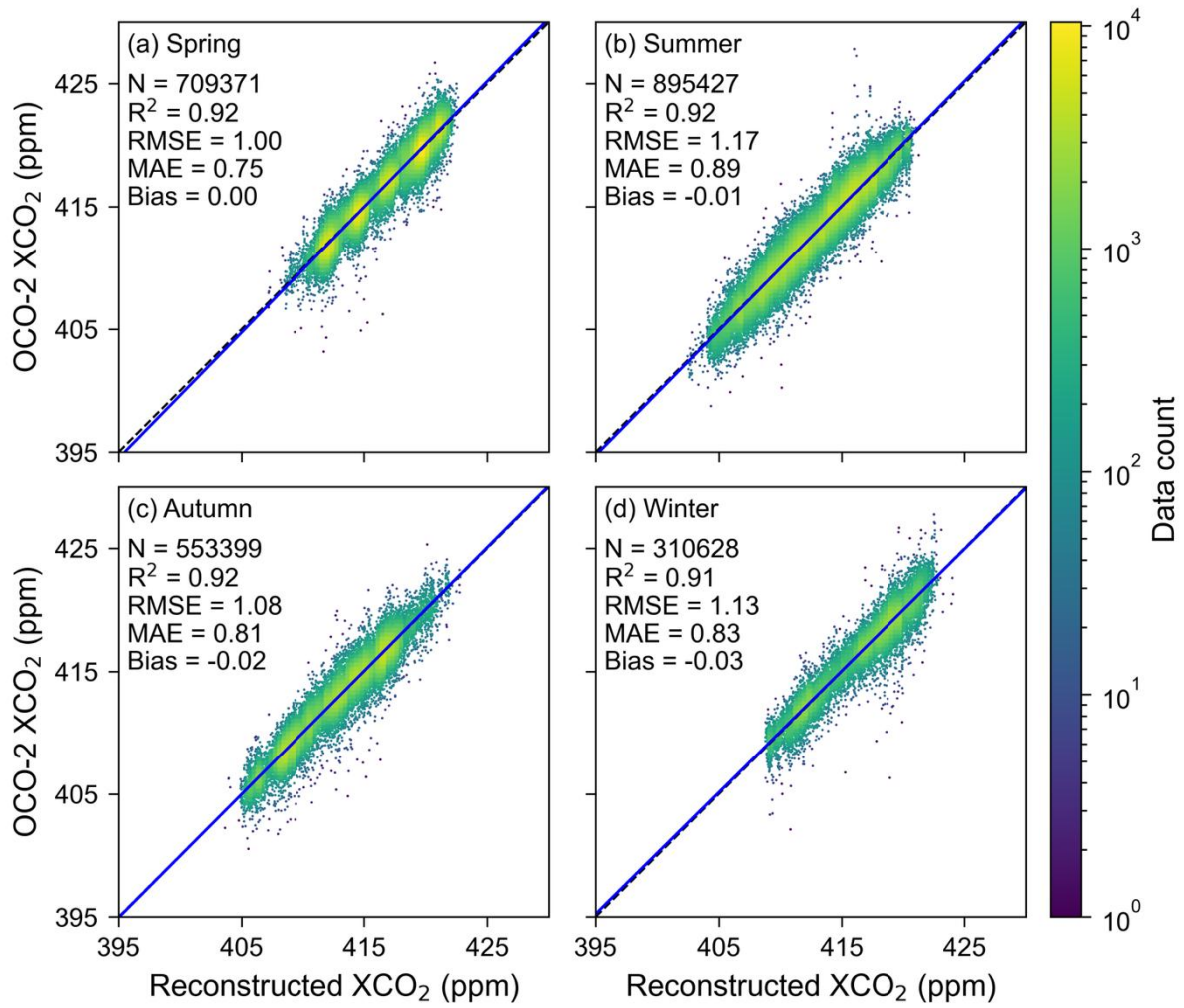


Figure S7. Comparison of our reconstructed XCO₂ dataset with the ACOS-OCO-2 product XCO₂ per season³. The black dashed line indicates the 1:1 reference, and the blue solid line shows the best-fit linear regression. The data volume is represented by color shading (see color bar for count values).

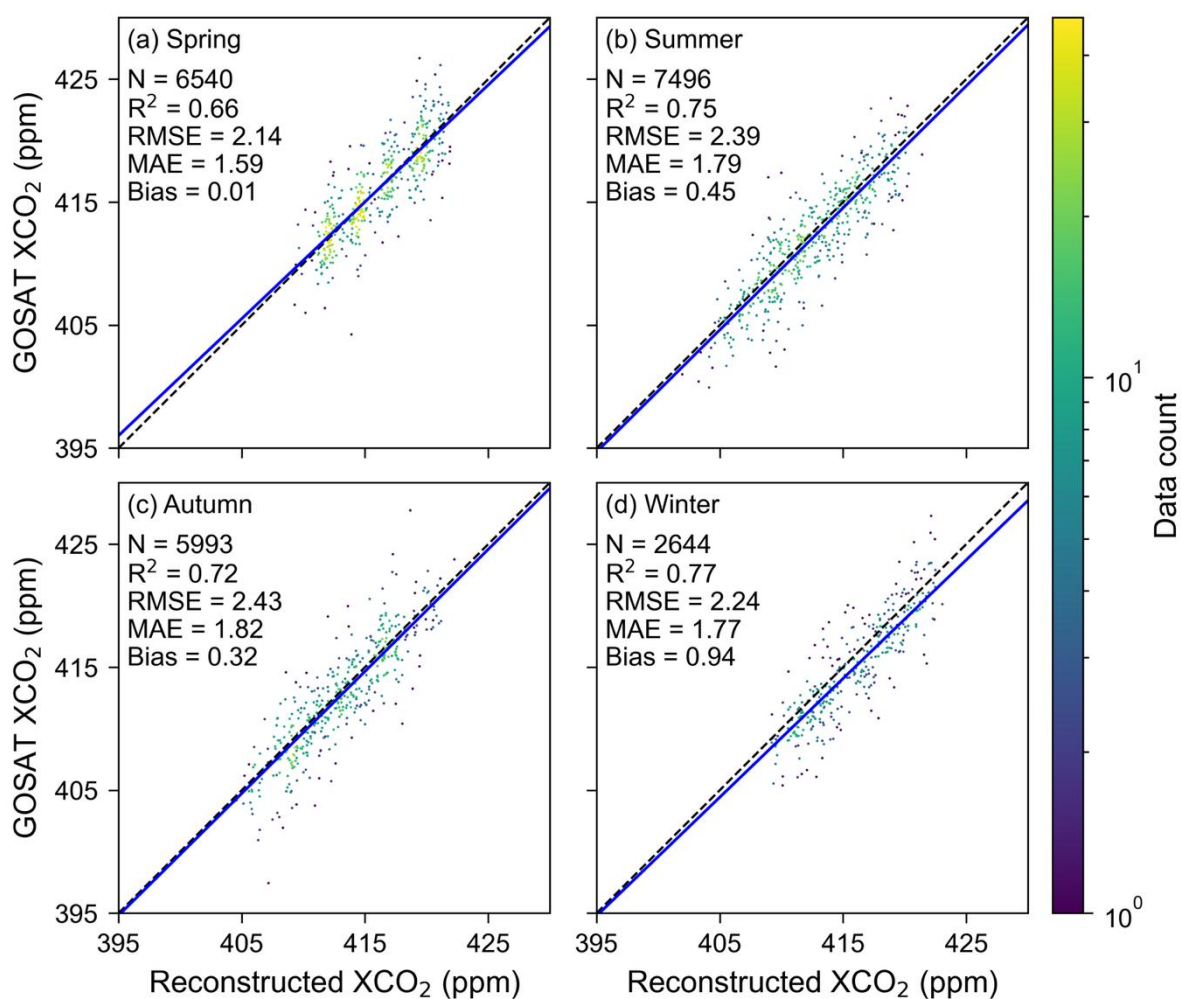


Figure S8. Comparison of our reconstructed XCO₂ dataset with the NIES-GOSAT product XCO₂ per season⁴. The black dashed line indicates the 1:1 reference, and the blue solid line shows the best-fit linear regression. The data volume is represented by color shading (see color bar for count values).

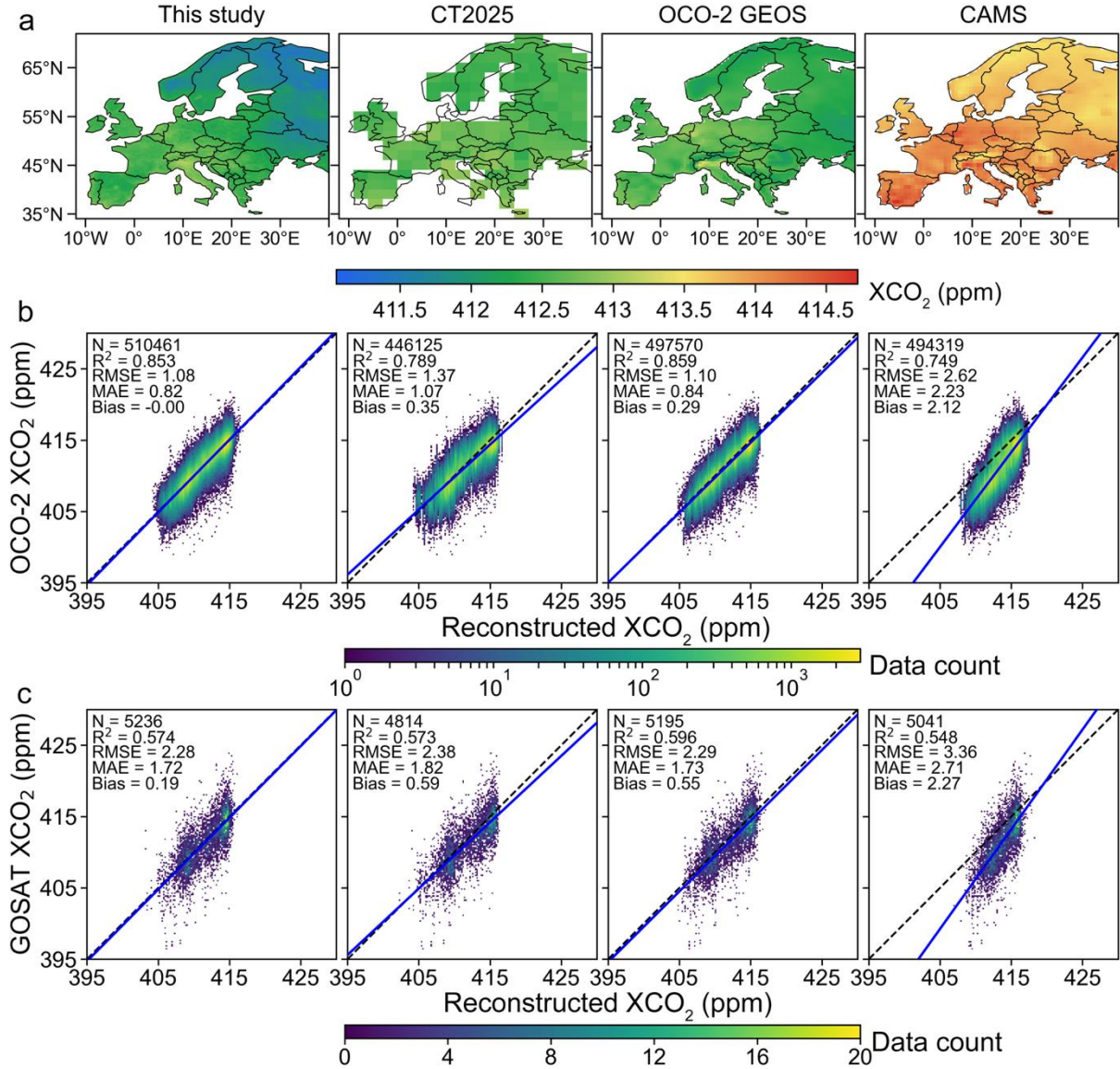


Figure S9. Comparison of the reconstructed XCO₂ dataset with other XCO₂ datasets in 2020. Panel (a) shows the annual mean spatial distributions in 2020 from this study, CT2025⁵, OCO-2 GEOS⁶, and CAMS^{7,8}. Panels (b) and (c) validate the four datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

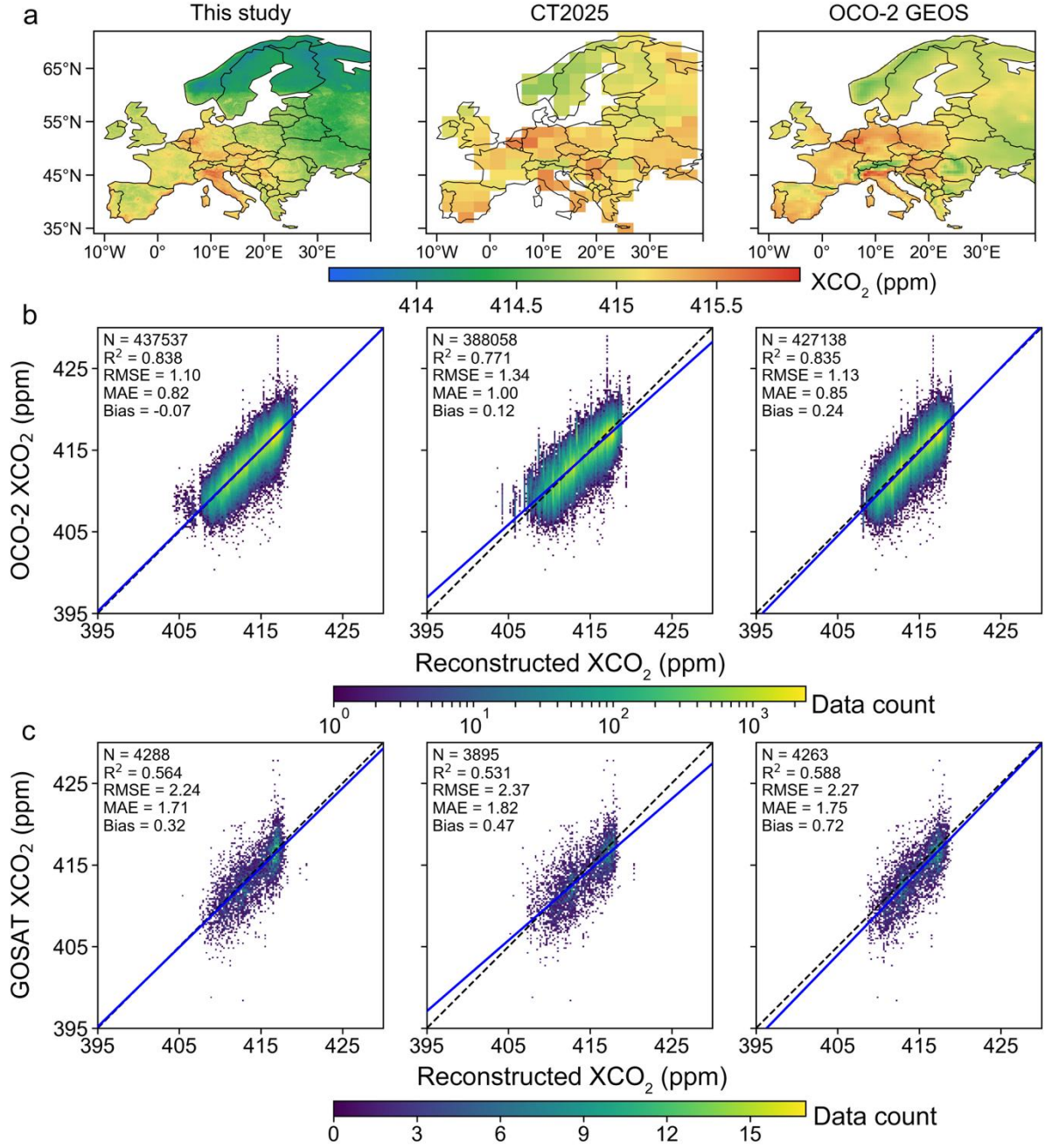


Figure S10. Comparison of the reconstructed XCO_2 dataset with other XCO_2 datasets in 2021. Panel (a) shows the annual mean spatial distributions in 2021 from this study, CT2025, and OCO-2 GEOS. Panels (b) and (c) validate the three datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

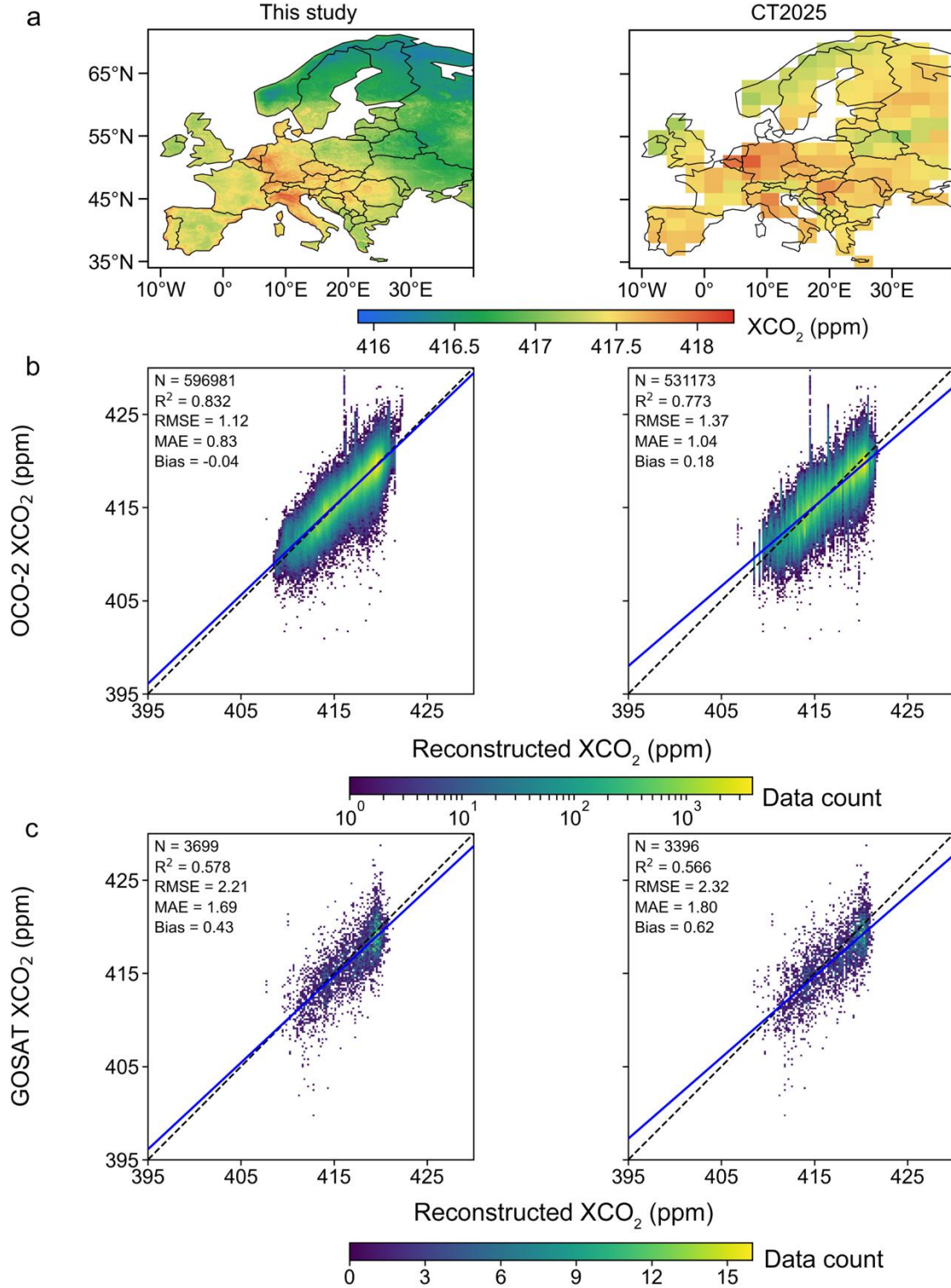


Figure S11. Comparison of the reconstructed XCO₂ dataset with other XCO₂ datasets in 2022. Panel (a) shows the annual mean spatial distributions in 2022 from this study and CT2025. Panels (b) and (c) validate the two datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

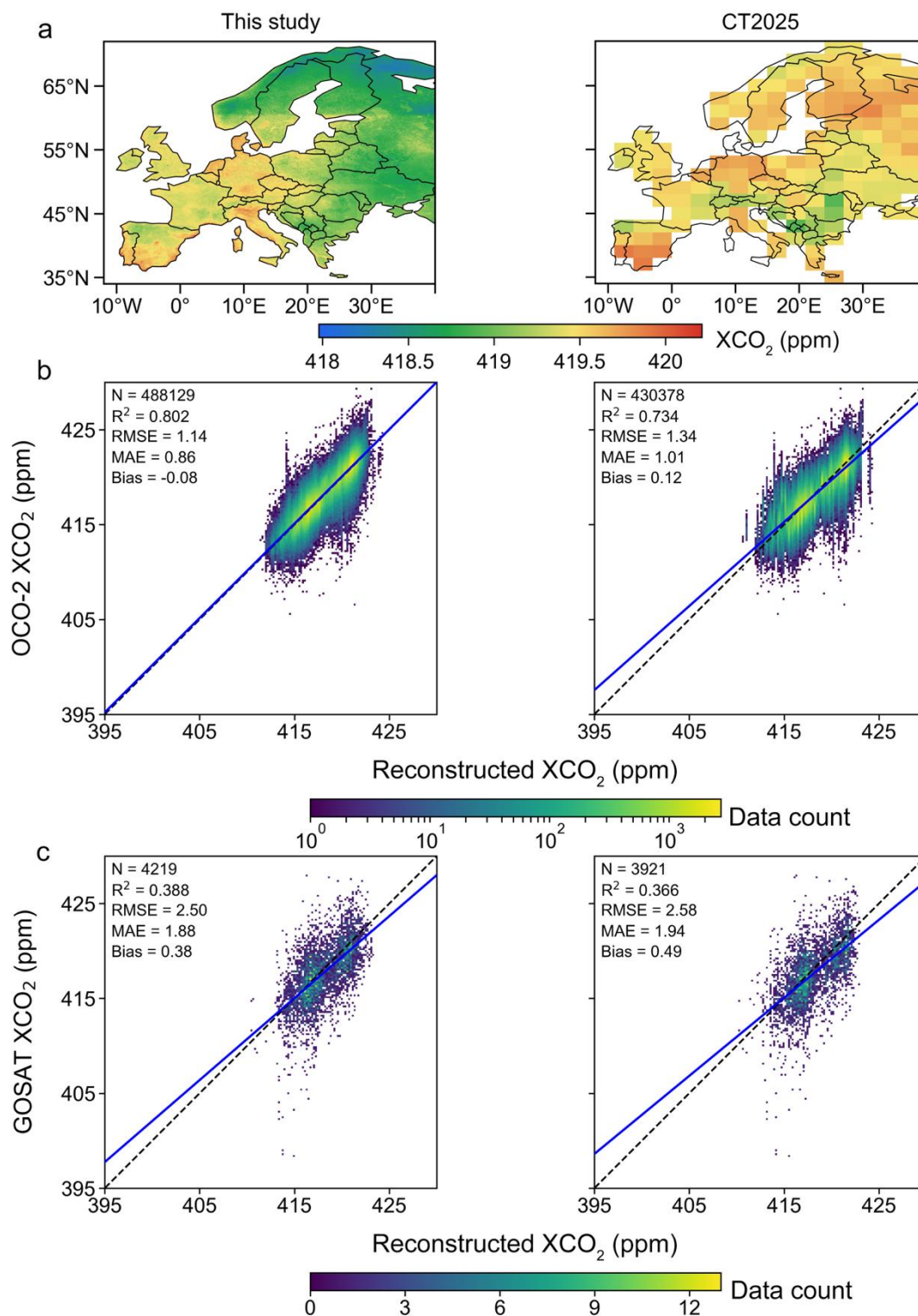


Figure S12. Comparison of the reconstructed XCO_2 dataset with other XCO_2 datasets in 2023. Panel (a) shows the annual mean spatial distributions in 2023 from this study and CT2025. Panels (b) and (c) validate the two datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

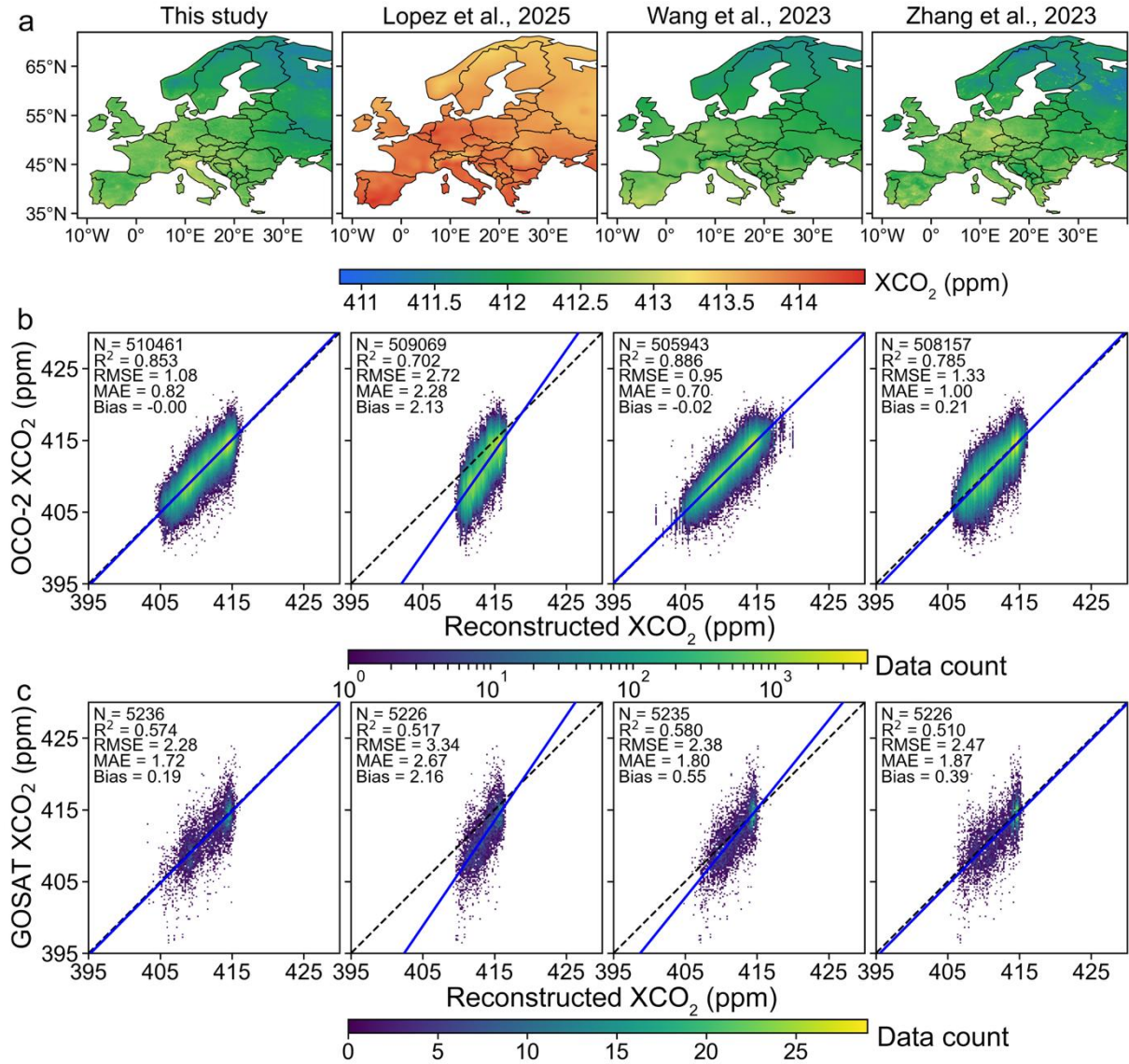


Figure S13. Comparison of the reconstructed XCO₂ dataset with peer-produced XCO₂ datasets in 2020. Panel (a) shows the annual mean spatial distributions in 2020 from this study, Lopez et al.⁹, Wang et al.¹⁰, and Zhang et al.¹¹. Panels (b) and (c) validate the four datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

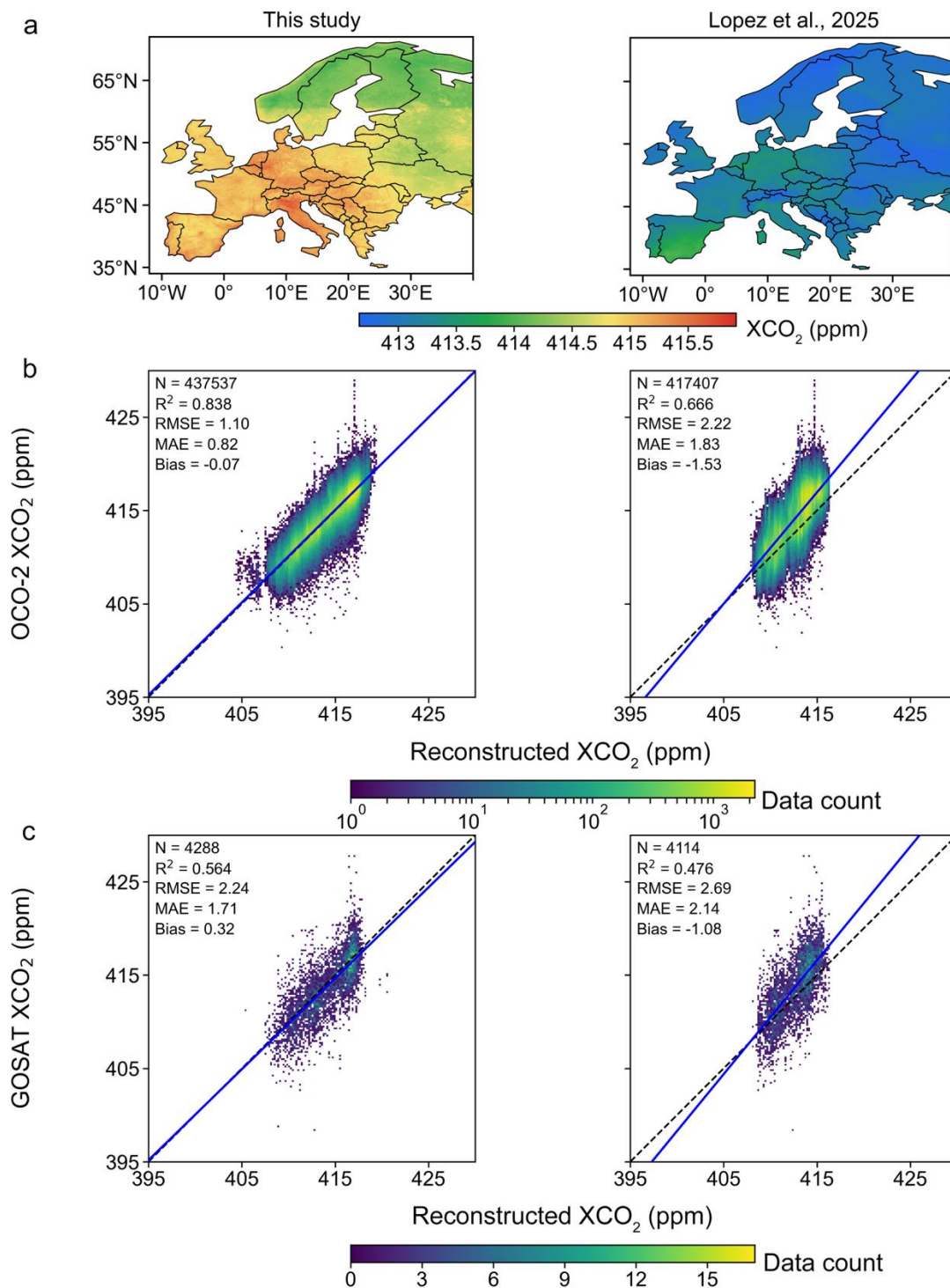


Figure S14. Comparison of the reconstructed XCO₂ dataset with peer-produced XCO₂ datasets in 2021. Panel (a) shows the annual mean spatial distributions in 2021 from this study and Lopez et al.⁹ Panels (b) and (c) validate the two datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

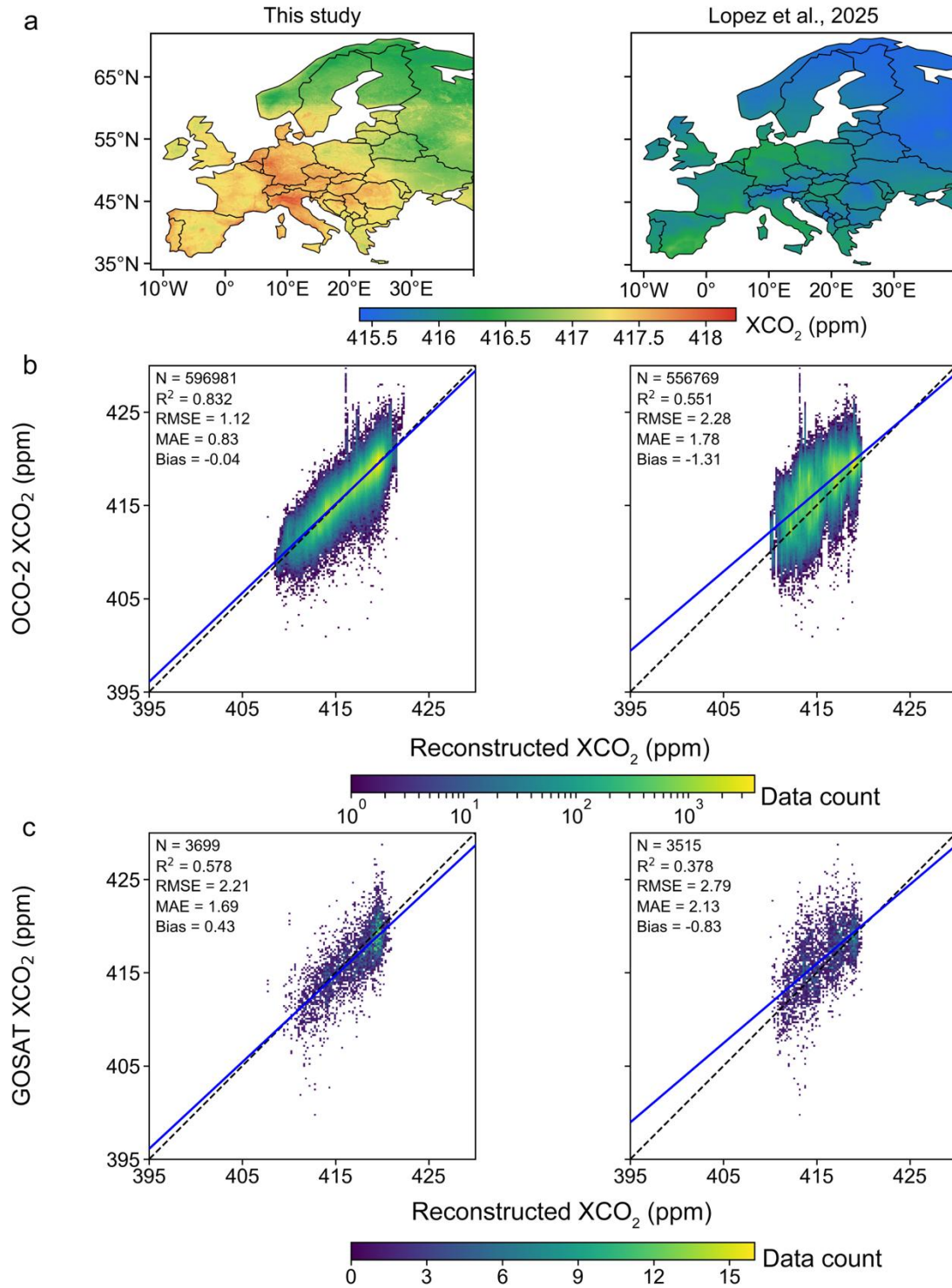


Figure S15. Comparison of the reconstructed XCO₂ dataset with peer-produced XCO₂ datasets in 2022. Panel (a) shows the annual mean spatial distributions in 2022 from this study and Lopez et al.⁹ Panels (b) and (c) validate the two datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

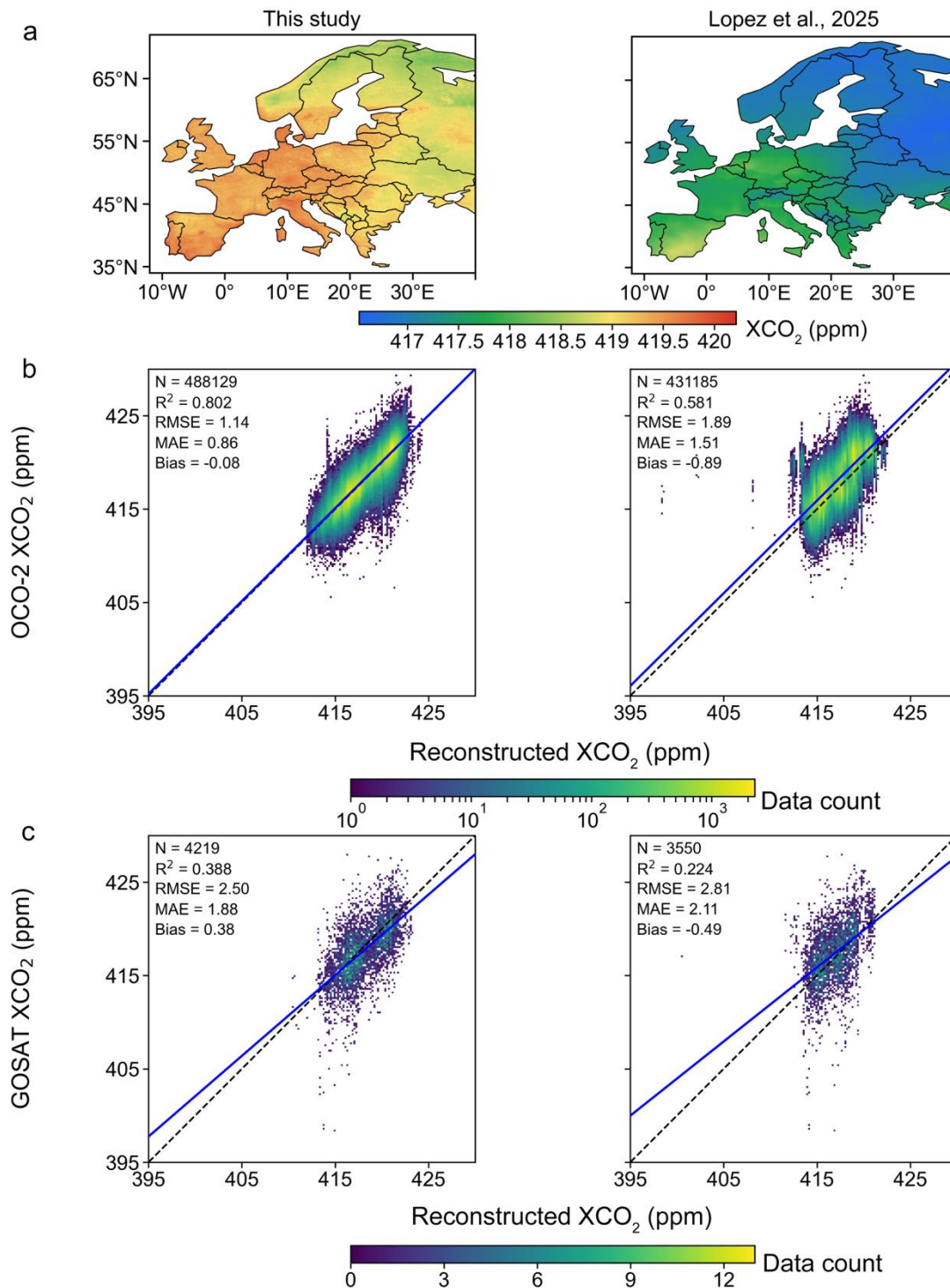


Figure S16. Comparison of the reconstructed XCO₂ dataset with peer-produced XCO₂ datasets in 2023. Panel (a) shows the annual mean spatial distributions in 2023 from this study and Lopez et al.⁹ Panels (b) and (c) validate the two datasets against the ACOS-OCO-2 and NIES-GOSAT products, respectively. Colors indicate data density, the black dashed lines denote the 1:1 relationship, and the blue solid lines represent the best-fit linear regressions.

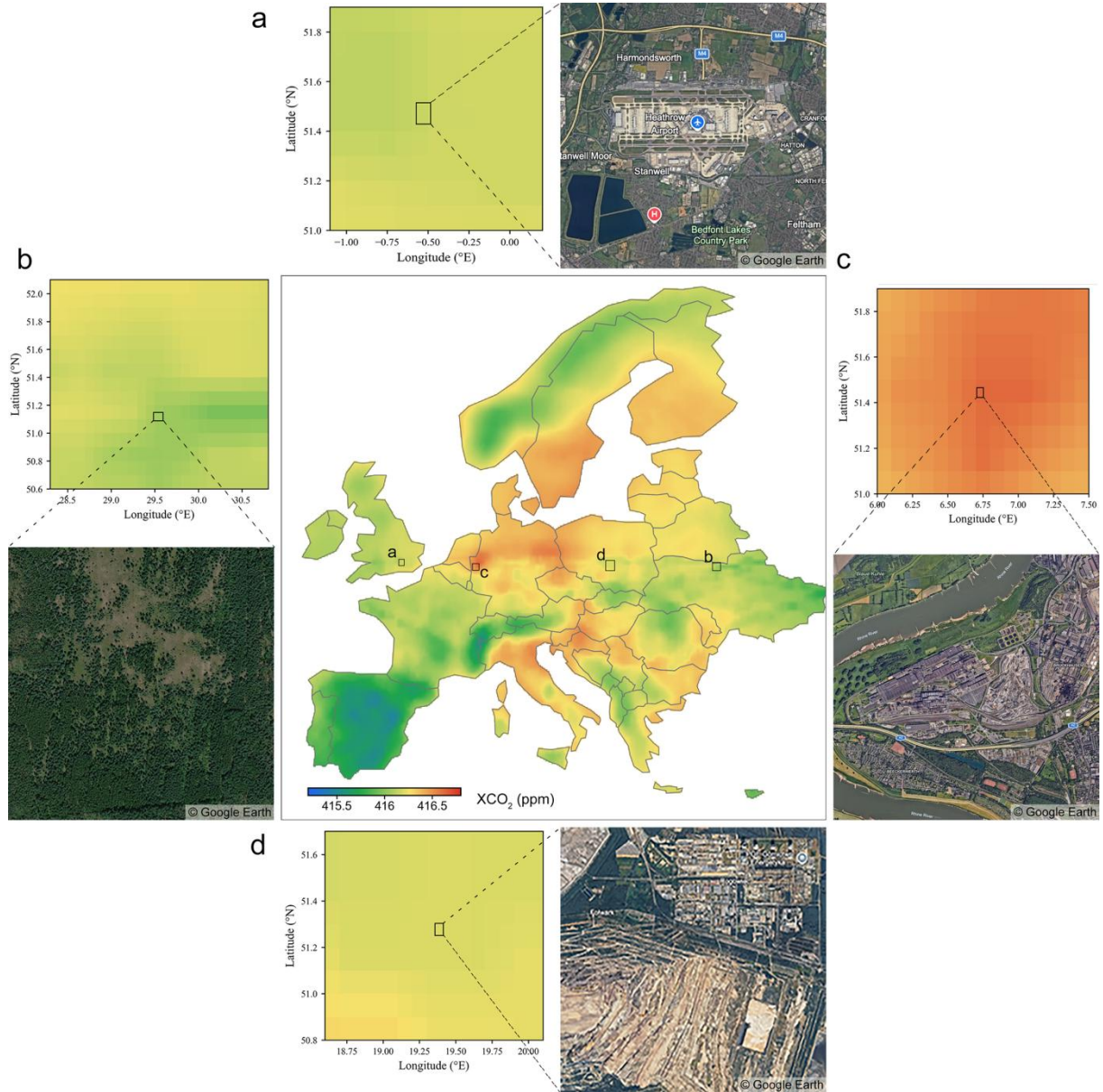


Figure S17. Examples of localized high-XCO₂ regions in the Lopez et al.⁹ dataset on April 8, 2020, showing (a) Heathrow Airport, United Kingdom, (b) the Chernobyl fires, Ukraine, (c) the Duisburg Steel Plant, Germany, and (d) the Bełchatów Power Station, Poland. Each panel displays the reconstructed daily XCO₂ concentration and the corresponding true color high-resolution satellite image from © Google Earth of the same location. The central map shows the locations of each selected example across terrestrial Europe.

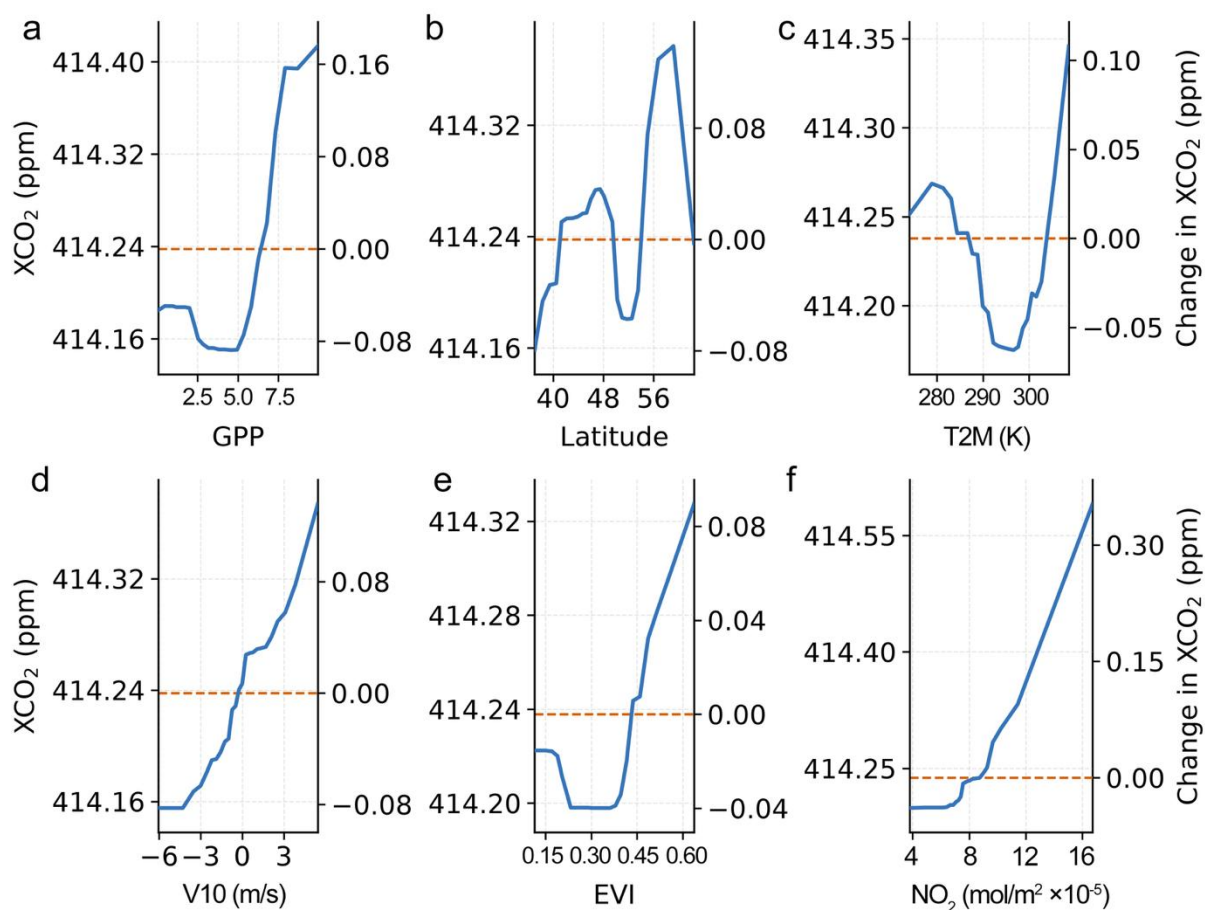


Figure S18. Partial Dependence Plots (PDPs) showing the marginal effects of predictors on our reconstructed XCO₂ dataset.¹² (a) Gross Primary Production (GPP), (b) Latitude, (c) 2-metre air temperature (T2M), (d) 10-metre northward wind component (V10), (e) Enhanced Vegetation Index (EVI), and (f) Nitrogen Dioxide (NO₂).

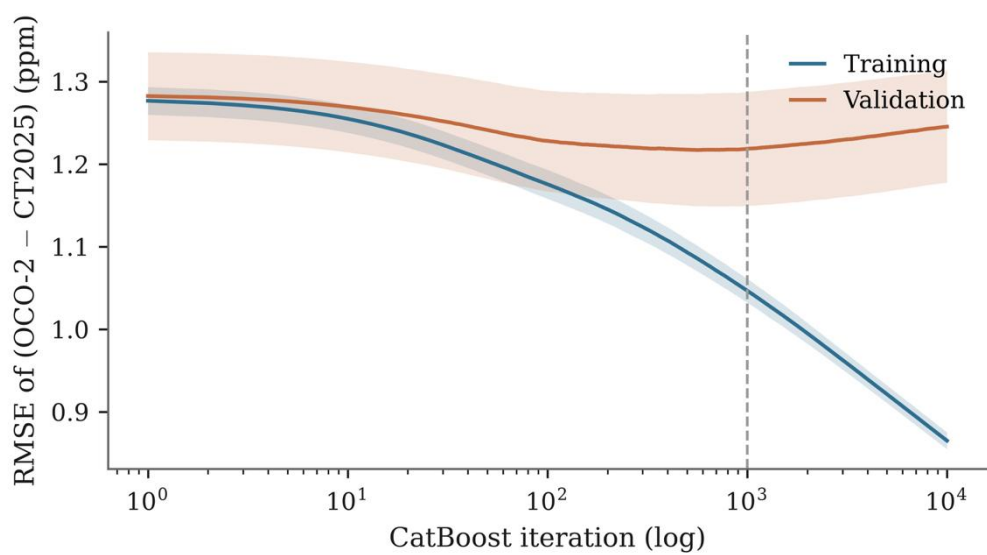


Figure S19. Mean training and validation RMSE of the CatBoost model across 10,000 iterations on a logarithmic scale.

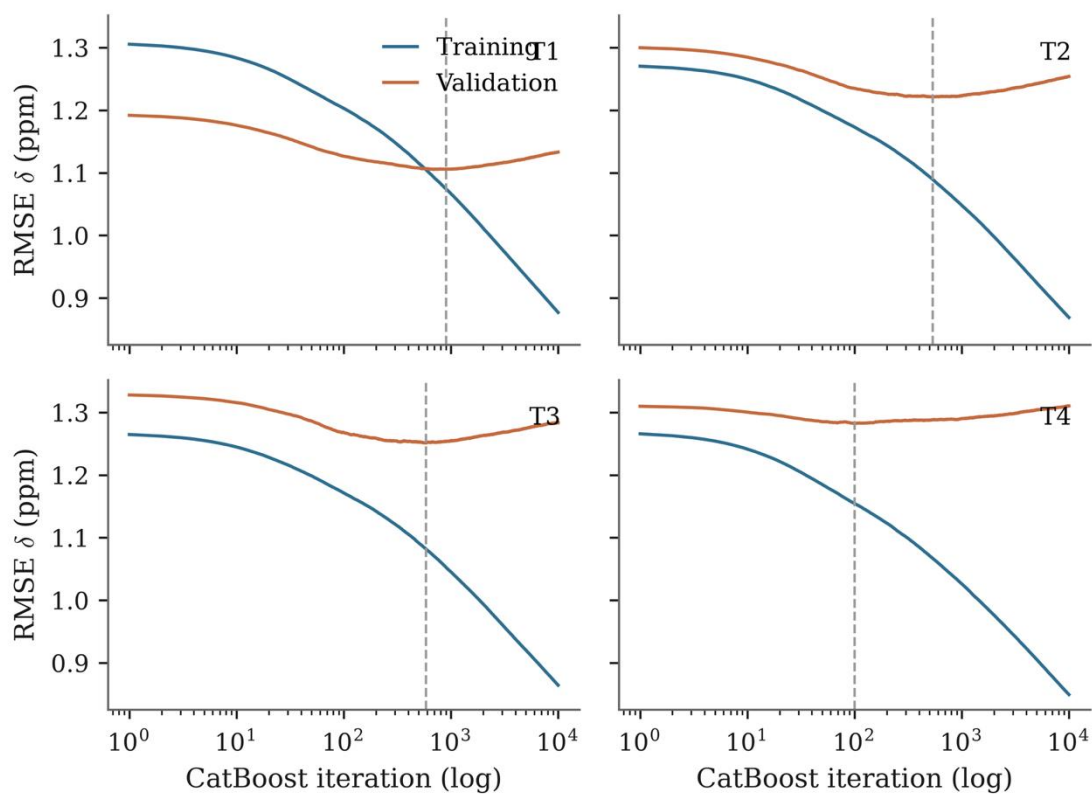


Figure S20. Individual training and validation RMSE trajectories across 10,000 iterations for temporal folds T1, T2, T3, and T4.

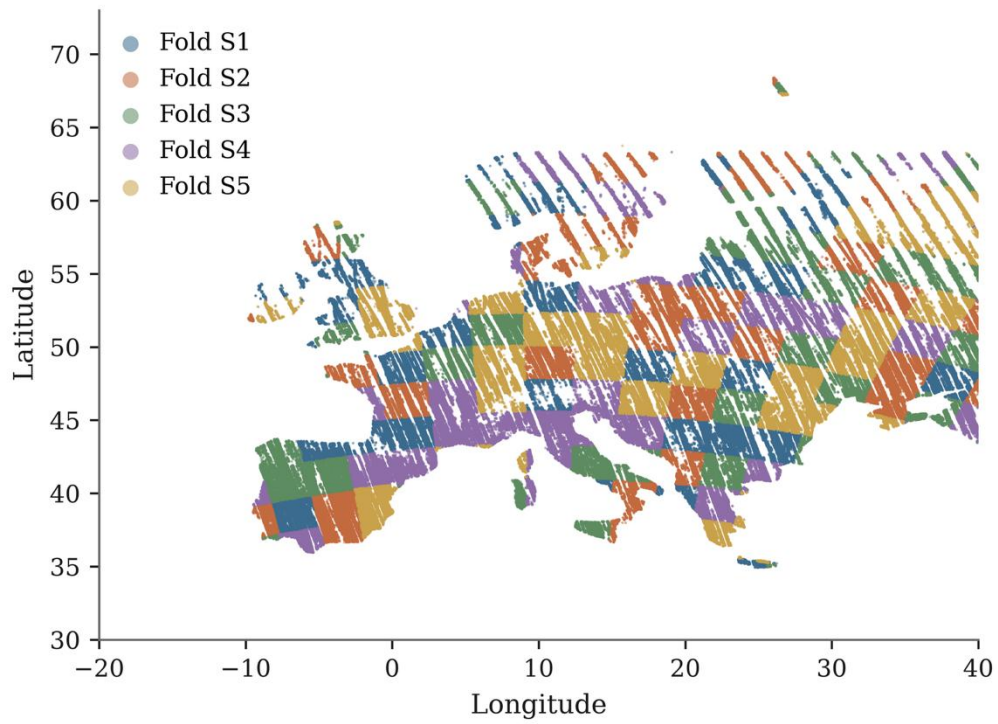


Figure S21. Spatial block assignment across 5 equal-area spatial folds (S1 to S5) over the European study area.

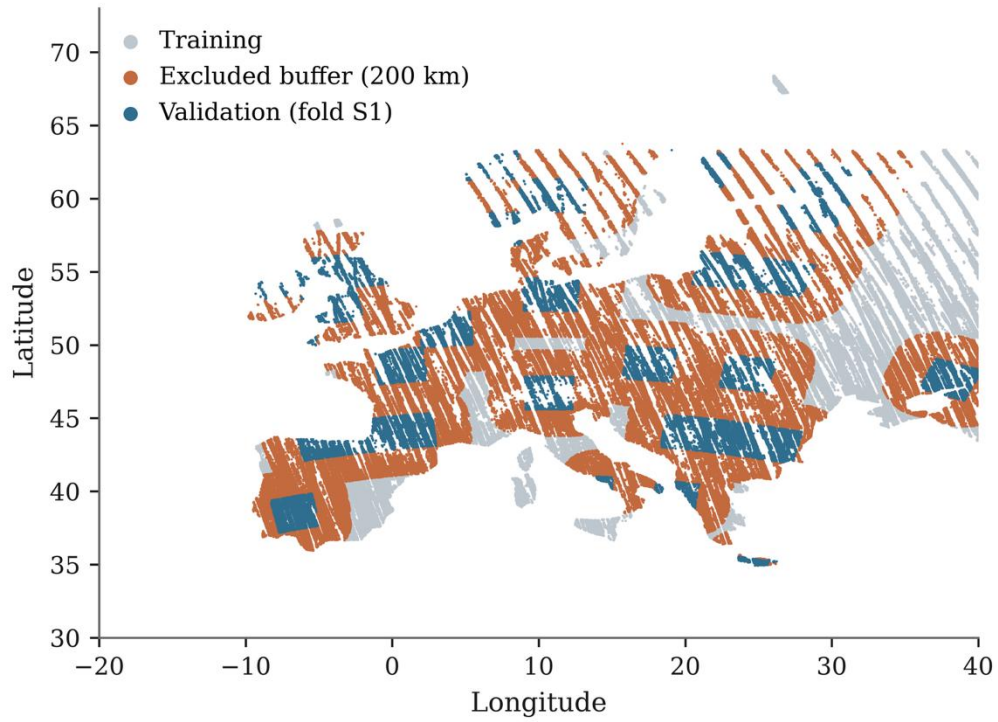


Figure S22. Spatial distribution of active validation blocks, adjacent training blocks, and the excluded 200 km buffer zone for spatial fold S1 across Europe.

Table S1. The information on the TCCON stations.

Name	ID	Latitude	Longitude	Country	Start Date	End Date*	Reference
Bremen	br	53.10°	8.85°	Germany	2009.01.06	2021.06.24	Notholt et al. ¹³
Garmisch	gm	47.48°	11.06°	Germany	2007.07.18	2023.05.04	Sussmann and Rettinger ¹⁴
Karlsruhe	ka	49.10°	8.44°	Germany	2014.01.15	2023.06.26	Hase et al. ¹⁵
Orleans	or	47.97°	2.11°	France	2009.09.06	2023.07.17	Warneke et al. ¹⁶
Paris	pr	48.85°	2.36°	France	2014.09.23	2023.12.20	Té et al. ¹⁷
Sodankyla	so	67.37°	26.63°	Finland	2009.05.16	2023.05.30	Kivi et al. ¹⁸

* The End Date does not indicate the cessation of station operations but denotes the last date of data used in this study.

Table S2. Detailed information on the variables used in this study.

Data type	Data name/Product name	Version	Variable	Abbreviation	Temporal resolution	Spatial resolution	Unit	Start date	End date	Source
Target variable	ACOS-OCO-2	L2_Lite_FP_v11.2r	XCO ₂	OCO-2	daily	2.25 km × 1.29 km	ppm	2014.09.06	2025.06.30	NASA
	NOAA CarbonTracker 2025	XCO ₂ mole CT2025/ mole	XCO ₂	CT2025	daily	3° × 2°	ppm	2000.01.01	2025.01.11	NOAA
	fraction product at 13:30 local solar time		XCO ₂ _1330LST							
Meteorological data	ERA5 hourly data on single levels from ERA5		10-metre eastward wind component	U10	hourly	0.25° × 0.25°	m·s ⁻¹	1940.01.01	2025.08.08	ECMWF
	1940 to present ¹⁹		10-metre northward wind component	V10	hourly	0.25° × 0.25°	m·s ⁻¹	1940.01.01	2025.08.08	ECMWF
			2-metre air temperature	T2M	hourly	0.25° × 0.25°	K	1940.01.01	2025.08.08	ECMWF
			Surface pressure	SP	hourly	0.25° × 0.25°	Pa	1940.01.01	2025.08.08	ECMWF
			Boundary layer height	BLH	hourly	0.25° × 0.25°	m	1940.01.01	2025.08.08	ECMWF
			2-metre dewpoint temperature	D2M	hourly	0.25° × 0.25°	K	1940.01.01	2025.08.08	ECMWF
			Total precipitation	TP	hourly	0.25° × 0.25°	m	1940.01.01	2025.08.08	ECMWF
	Post-processing calculations (Use ERA5)	Calculate	Relative humidity	RH	daily	0.25° × 0.25°	%	2019.01.01	2023.12.31	/
			Wind speed	WS	daily	0.25° × 0.25°	m·s ⁻¹	2019.01.01	2023.12.31	/
Land surface data	MODIS/Terra Vegetation Indices 16-Day	MOD13A2 V061	1_km_16_days_EVI,	EVI	16 days	1 km × 1 km	/	2000.02.18	2025.08.14	NASA
	L3 Global 1km SIN Grid V061 ²⁰		1_km_16_days_VI_Quality,							
			1_km_16_days_pixel_reliability							
	MODIS/Terra+Aqua Land Cover Type	MCD12Q1 V061	LC_Type3	LC	yearly	500 m × 500 m	/	2001	2025	NASA
	Yearly L3 Global 500m SIN Grid V061 ²¹									
	MODIS/Terra Snow Cover Daily L3 Global	MOD10A1 V061	NDSI_Snow_Cover	NDSI	daily	500 m × 500 m	/	2000.01.24	2025.08.14	NSIDC
	500m SIN Grid V061 ²²		NDSI_Snow_Cover_Basic_QA							
	Gross Primary Production ²³	GPP_500M_V60	GPP	GPP	daily	500 m × 500 m	g C·m ⁻²	2000.02.18	2024.12.26	HKUgeography
Anthropogenic source data	Global daily emissions for each file at 1 km resolution, the base and speciated VOCs based on MODISVIIRS data ²⁴		FINN v2.5	CO ₂	daily	1 km × 1 km	mol	2012.01.01	2023.12.31	NCAR
			MODISVIIRS	AREA						
	ODIAC Fossil fuel emission dataset ²⁵		ODIAC2024	FFCO ₂	monthly	1 km × 1 km	Tonne C	2000.01	2023.12	NIES
	Sentinel-5P OFFL NO ₂ : Offline Nitrogen Dioxide ²⁶	Sentinel-5P OFFL	NO ₂ _column_number_density	NO ₂	daily	1113.2 m × 1113.2 m	mol·m ⁻²	2018.06.28	2025.10.20	Google Earth Engine
	VNP46A2: VIIRS Lunar Gap-Filled BRDF Nighttime Lights Daily L3 Global 500m ²⁷	VNP46A2	Gap_Filled_DNB_BRDF_Corrected_NTL	NTL	daily	500 m × 500 m	/	2012.01.19	2025.12.26	NASA
Evaluation data	Total Carbon Column Observing Network	GGG2020	XCO ₂	TCCON	30 mins	point	ppm	each point different		Different
	NIES-GOSAT	v03.05_Level2	XCO2BiasCorrected	NIES-GOSAT	daily	10.5 km × 10.5km	ppm	2009.04.13	2024.11.30	NIES
			totalPreScreeningResult							
			totalPostScreeningResult							

Table S3. Hyperparameter search spaces for the CatBoost model and candidate residual-correction methods. The best parameters are shown in bold.

(a) CatBoost model		
Hyperparameter	Description	Search Space / Configuration
loss_function	Objective function to minimize	Fixed: "RMSE"
iterations	Maximum number of trees (estimators) spanning multiple orders of magnitude	[10, 100, 1000 , 5000, 10000]
learning_rate	Step size shrinkage used in updates	[0.005, 0.01, 0.02 , 0.03, 0.05, 0.1]
depth	Depth of the decision trees	[4, 6 , 8, 10]
l2_leaf_reg	L2 regularization coefficient on leaf values	[1, 3 , 5, 10, 20]
random_strength	Score standard deviation multiplier for random split selection	[0 , 1, 2]
bootstrap_type	Method used for sampling weights	["Bernoulli", " Bayesian "]
subsample	Sample rate for bagging (active only when bootstrap_type is Bernoulli)	[0.6, 0.8, 1.0]
bagging_temperature	Intensity of Bayesian bootstrapping (active only when bootstrap_type is Bayesian)	[0, 0.5, 1.0]
random_seed	Seed to ensure reproducibility of training	Fixed: 42
task_type	Hardware target utilized for model training	Fixed: " CPU "
(b) Inverse distance weighting (IDW)		
Parameter	Description	Search Space
power	Exponent parameter controlling the weight decay with distance	[1 , 2, 3]
radius_km	Maximum search radius in kilometers	[50 , 100, 200, 300, 500]
k_neighbors	Number of nearest neighbors used for interpolation	[5, 10, 20, 50]
time_window	Temporal search window for finding daily residuals	[" same_day ", "+/-1 day", "+/-3 days", "+/-7 days"]
lam	Scaling/shrinkage coefficient (λ) for the residual correction	[0.25 , 0.5, 0.75, 1.0]
(c) Ordinary kriging (OK)		
variogram_model	Theoretical variogram model used to fit spatial covariance	[" spherical ", "exponential", "gaussian", "matern"]
range_km	Correlation range of the variogram model in kilometers	[100 , 200, 300, 500, "auto"]
nugget	Constraint on the nugget effect representing small scale noise	[" auto ", "constrained"]
k_neighbors	Number of nearest neighbors used for local kriging	[20, 50, 100]
radius_km	Search radius in kilometers	[200, 500, 800]

time_window	Temporal search window for finding daily residuals	["same_day", "+/-1 day", " +/-3 days"]
lam	λ for the residual correction	[0.25 , 0.5, 0.75, 1.0]
(d) RF-assisted regression kriging (RK)		
secondary	Machine learning algorithm used to fit the deterministic trend	["ridge", "rf_shallow" , "small_catboost"]
variogram_model	Theoretical variogram model used for the residuals of the trend	["spherical", "exponential"]
range_km	Correlation range of the variogram in kilometers	[200, 500, "auto"]
k_neighbors	Number of nearest neighbors used for local kriging	[20, 50, 100]
radius_km	Search radius in kilometers	[200, 500, 800]
time_window	Temporal search window for finding daily residuals	["same_day", " +/-3 days"]
lam	λ for the residual correction	[0.25 , 0.5, 0.75, 1.0]
(e) Spatiotemporal Gaussian-process correction (GP)		
kernel	Covariance kernel function used to model spatiotemporal correlation	["RBF", "Matern"]
spatial_length_scale_km	Spatial correlation length scale in kilometers	[50 , 100, 200, 500]
temporal_length_scale_day	Temporal correlation length scale in days	[1 , 3, 7]
noise_level	Standard deviation of the observation noise	[0.1, 0.3, 0.5, 1.0]
lam	λ for the residual correction	[0.25 , 0.5, 0.75, 1.0]

Table S4. Temporal-validation sensitivity results for the candidate model configurations.

Model configuration	R ²	RMSE (ppm)	MAE (ppm)	Bias (ppm)
CatBoost	0.81	1.22	0.90	0.01
CatBoost + IDW	0.81	1.22	0.90	0.01
CatBoost + OK	0.81	1.23	0.91	0.03
CatBoost + RK	0.81	1.23	0.91	0.03
CatBoost + GP	0.81	1.22	0.90	0.01

Table S5. Detailed fold-level performance metrics of the 4-fold temporal leave-one-year-out CV (T1–T4).

Fold	Train Year	Validation Year	Train Samples	Validation Samples	Train R^2	Val R^2	Train RMSE (ppm)	Val RMSE (ppm)	Train MAE (ppm)	Val MAE (ppm)	Train Bias (ppm)	Val Bias (ppm)
T1	2020, 2021, 2022	2019	487530	149299	0.91	0.83	1.07	1.11	0.78	0.83	0.00	0.23
T2	2019, 2021, 2022	2020	471178	165651	0.94	0.82	1.05	1.22	0.77	0.91	0.00	-0.02
T3	2019, 2020, 2022	2021	495241	141588	0.94	0.80	1.05	1.25	0.77	0.93	0.00	-0.16
T4	2019, 2020, 2021	2022	456538	180291	0.91	0.80	1.03	1.29	0.75	0.94	0.00	-0.03
Mean					0.92	0.81	1.05	1.22	0.77	0.90	0.00	0.01

Table S6. Detailed fold-level performance metrics of the 5-fold spatial blocked CV under 200 km buffer constraint (S1–S5).

Fold	Train Samples	Validation Samples	Median Distance (km)	Train R^2	Val R^2	Train RMSE (ppm)	Val RMSE (ppm)	Train MAE (ppm)	Val MAE (ppm)	Train Bias (ppm)	Val Bias (ppm)
S1	147,005	127,367	278.43	0.94	0.90	0.98	1.19	0.71	0.89	0.00	-0.08
S2	173,760	127,366	281.16	0.94	0.91	1.01	1.18	0.74	0.86	0.00	0.04
S3	179,767	127,366	294.95	0.94	0.91	1.02	1.15	0.74	0.86	0.00	0.02
S4	172,407	127,365	282.92	0.94	0.91	0.99	1.21	0.73	0.90	0.00	0.00
S5	217,935	127,365	294.22	0.93	0.90	1.00	1.26	0.72	0.93	0.00	-0.07
Mean				0.94	0.91	1.00	1.20	0.73	0.89	0.00	-0.02

Table S7. Performance during CV and independent testing.

	N	R ²	RMSE (ppm)	MAE (ppm)	Bias (ppm)
Temporal CV	636,829	0.91	1.22	0.91	0.01
Spatial CV	636,829	0.91	1.20	0.89	−0.02
Independent 2023 test	153,500	0.76	1.30	0.98	−0.13

Text S1. Sensitivity analysis of residual-correction methods and model selection

To evaluate whether spatiotemporal residual correction improves the CatBoost predictions, we tested four alternative methods: IDW²⁹, OK³⁰, RK³¹, and GP³². Hyperparameters were optimized only within the 2019–2022 development set. The hyperparameter search spaces for CatBoost and the candidate correction methods are provided in Table S3. The temporal CV results show that adding these correction terms produces no obvious improvement over CatBoost alone (Table S4). Therefore, according to the principle of parsimony, we retain CatBoost alone as the final model without post-hoc residual correction. The final CatBoost configuration consisted of 1,000 iterations, a learning rate of 0.02, a depth of 6, an L2 leaf regularization coefficient of 3.0, a random strength of 0.0, and Bayesian bootstrap with a bagging temperature of 1.0.

Text S2. Spatiotemporal CV performance metrics

Temporal CV used four leave-one-year-out folds (T1 to T4) over the development set 2019–2022. In each fold, one complete year was used for validation, and the other three years were used for training. Training convergence was monitored up to 10,000 iterations, and the validation error stabilized near 1,000 iterations (Figures S19 and S20). The reported mean statistics were calculated by averaging the validation results from the four folds (Table S5).

The spatial CV of the European study domain was divided into 250 km × 250 km equal-area square blocks, and each block was randomly assigned to one of five spatial folds (S1 to S5; Figure S21). When one fold was used for validation, all training samples within 200 km of the validation blocks were excluded (Figure S22). The fold-level training and validation results were provided in Table S6.

All 2023 data formed an independent test set and were completely held out during model development. The overall CV and independent test results are summarized in Table S7. The model maintained stable performance.

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