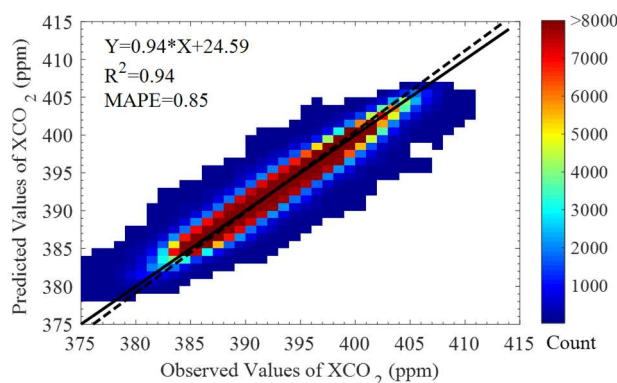


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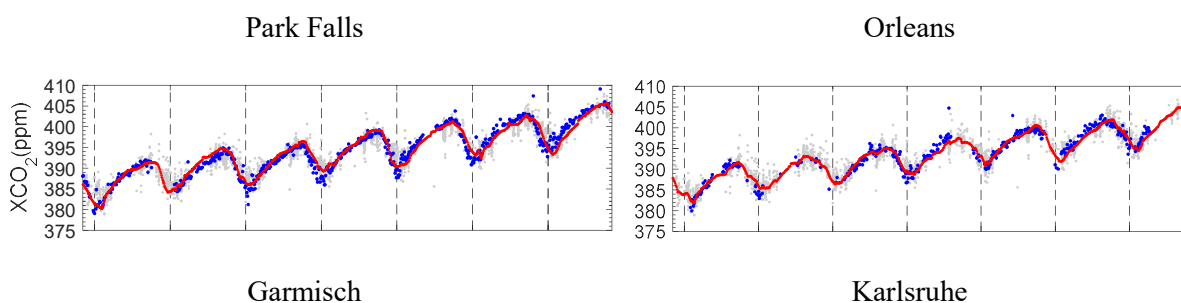
# Detection of Spatiotemporal Extreme Changes in Atmospheric CO<sub>2</sub> Concentration Based on Satellite Observations

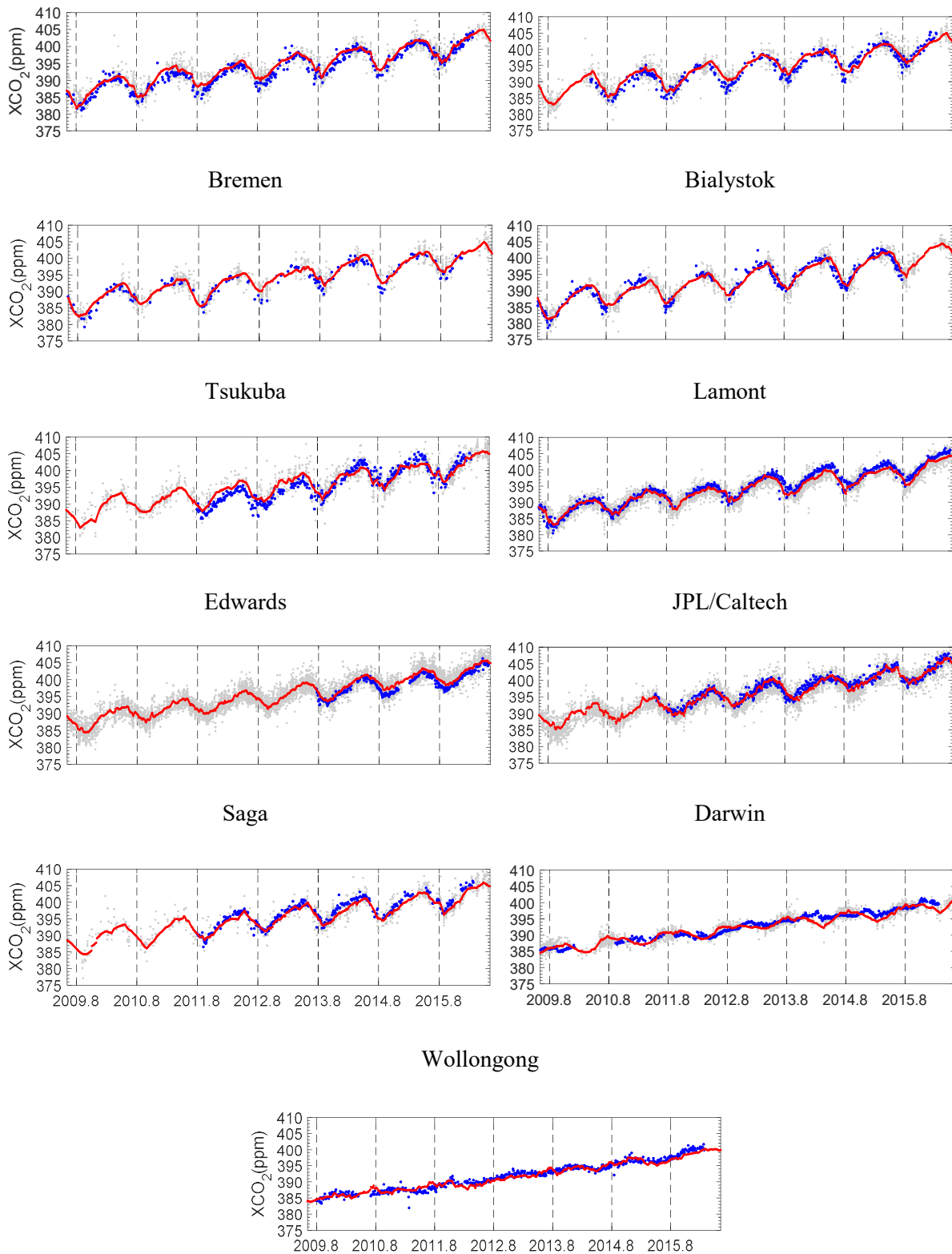
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**Figure S1.** The relationship between predicted XCO<sub>2</sub> and observed XCO<sub>2</sub> values in cross-validation of global land mapping of XCO<sub>2</sub>. The color grids represent the density of data distribution. The dotted line is derived from linear regression of predicted values of XCO<sub>2</sub> (Y) and the observed values of XCO<sub>2</sub> (X), which shows a significant linear relationship with R<sup>2</sup> equals 0.94 (p-value < 0.01) and good consistency of observed XCO<sub>2</sub> and predicted XCO<sub>2</sub> with mean absolute prediction error (MAPE) equal to 0.85. The solid line shows the one-to-one line.



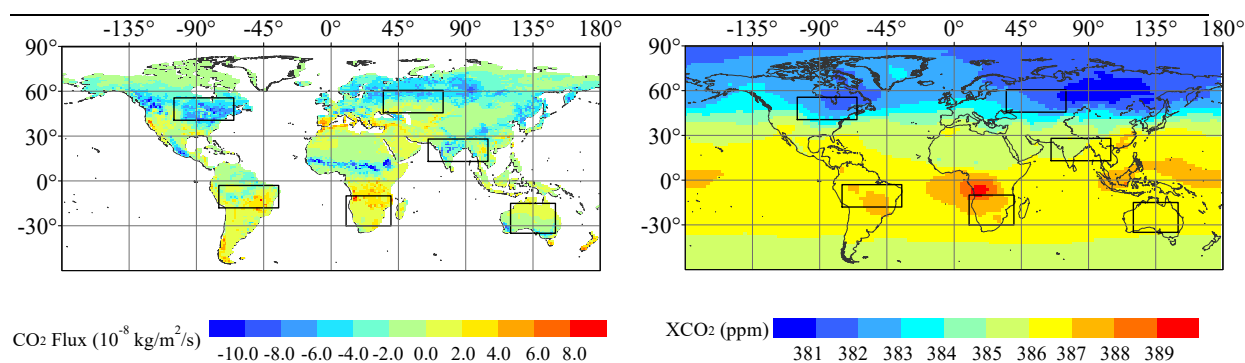


**Figure S2.** Temporal variation comparison for the 13 Total Carbon Column Observing Network (TCCON) sites. As shown in these panels, the original ACOS-GOSAT XCO<sub>2</sub> retrievals within 500 km of the TCCON site are in gray dots. The TCCON data, smoothed by applying the ACOS-GOSAT averaging kernel, are indicated by blue dots. The data are chosen using coincidence criteria of within  $\pm 2$  hours of GOSAT overpass time, and a 3-day (one time-unit) mean is calculated for the comparison. The predicted TCCON site XCO<sub>2</sub> time series using the

mapping approach are indicated by the red dots.

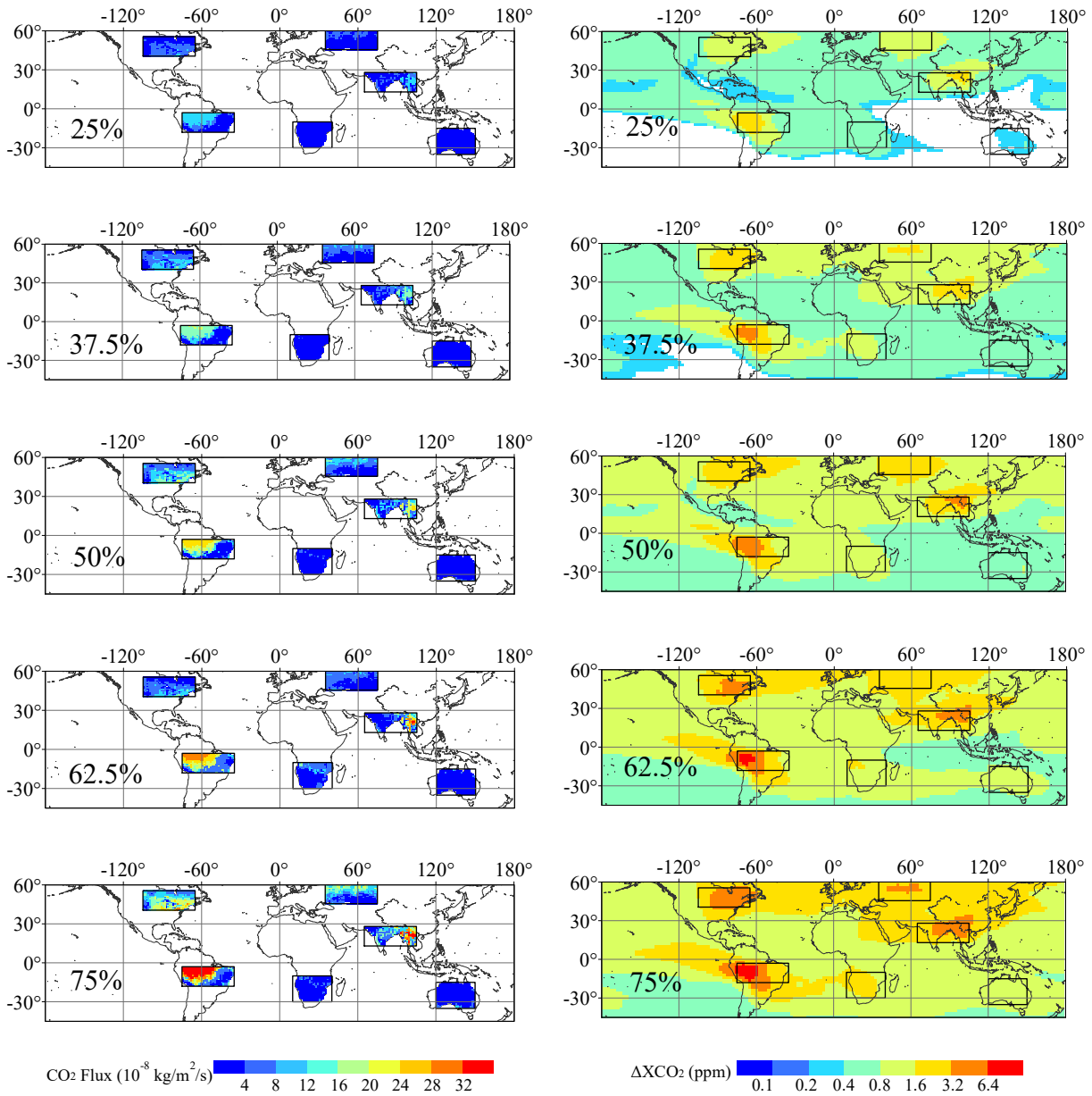
**Table S1.** Statistics of comparison between GM-XCO<sub>2</sub> and TCCON data (smoothed by applying the ACOS-GOSAT averaging kernel). Bias is calculated using GM-XCO<sub>2</sub> minus TCCON XCO<sub>2</sub> for each coincident data pair and averaged for each site.

Sites	Location (Latitude, longitude)	Coincident Data Pairs	Averaged Biases (ppm)	Averaged Absolute Bias (ppm)	Standard Deviation (ppm)
Bialystok	(53.23, 23.02)	377	0.09	0.96	1.68
Bremen	(53.1, 8.85)	191	0.67	1.15	1.75
Karlsruhe	(49.1, 8.44)	346	0.64	1.29	2.09
Orleans	(47.97, 2.11)	411	-0.05	0.89	1.41
Garmisch	(47.48, 11.06)	478	0.90	1.19	1.37
Park Falls	(45.94, -90.27)	665	0.11	1.02	1.76
Lamont	(36.6, -97.49)	761	-0.57	0.92	1.02
Tsukuba	(36.05, 140.12)	412	0.60	1.66	3.61
Edwards	(34.96 -117.88)	297	1.07	1.10	0.45
PL/Caltech	(34.2, -118.18)	584	-0.20	0.86	1.14
Saga	(33.24, 130.29)	329	-0.71	0.96	1.01
Darwin	(-12.43, 130.89)	612	-0.25	1.02	1.46
Wollongong	(-34.41, 150.88)	609	-0.19	0.77	0.93
Overall		6072	0.16	1.06	1.51



(a) Mean biosphere CO<sub>2</sub> fluxes from SiB3 over July 1 to September 31 of 2009

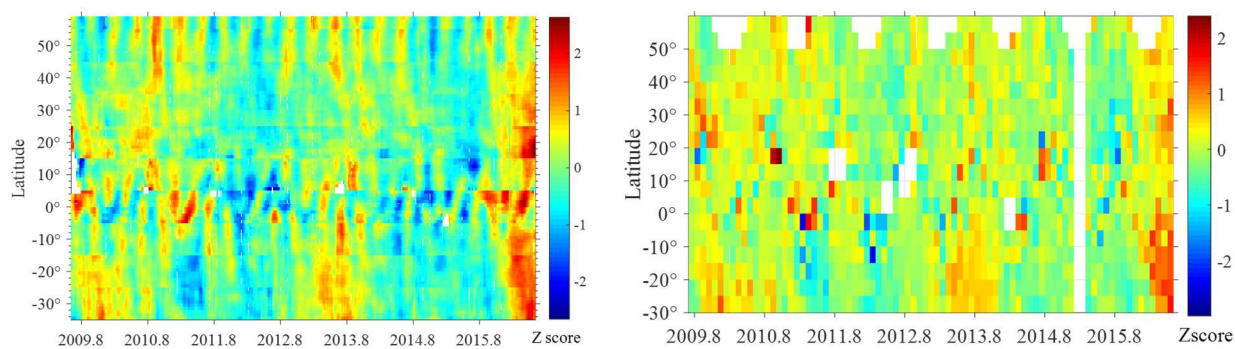
(b) Mean GEOS-XCO<sub>2</sub> using original SiB3 over July 1 to September 31 of 2009



(c) Different ratio (25%; 32.5%; 50%; 62.5%; 75%) of biospheric CO<sub>2</sub> fluxes as CO<sub>2</sub> emission

(d) Different GEOS-CO<sub>2</sub> response to different ratio of CO<sub>2</sub> fluxes change in (c).

**Figure S3.** Different biospheric CO<sub>2</sub> fluxes influence on XCO<sub>2</sub> from GEOS-Chem model simulation. They are mean biospheric CO<sub>2</sub> fluxes and corresponding XCO<sub>2</sub> output shown in (a) and (b). Different enhancements of local biospheric CO<sub>2</sub> fluxes as emission input for simulating different carbon sources/sinks changes are shown in (c). Different XCO<sub>2</sub> output for different CO<sub>2</sub> flux change are shown in (d).

(a) Z score of GM-XCO<sub>2</sub>(b) Z score of original ACOS-XCO<sub>2</sub>

**Figure S4.** Latitudinal-temporal Z score of XCO<sub>2</sub> fitting residuals from GM-XCO<sub>2</sub> (a) and original ACOS-XCO<sub>2</sub> (b).

Red represents high possibility of extreme highly increased XCO<sub>2</sub>.



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