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Supplementary information

Large Chinese land carbon sink estimated from atmospheric carbon dioxide data

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Supplementary Information (SI)

Large Chinese terrestrial carbon sink estimated from atmospheric CO_2 data

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The SI is organized into six sections:

1 Atmospheric CO_2 Data

NOAA/ESRL ObsPack database

We use discrete (weekly) flask air samples from 105 sites and continuous (hourly) observations from 52 sites that are part of the global atmospheric surface CO_2 observations network. These are currently described by the Observation Package (ObsPack) obspack_co2_1_GLOBALVIEWplus_v4.1_2018-10-29 data product (ObsPack, 2018) from National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory (ESRL). We use these data to estimate large-scale CO_2 fluxes that represent upwind boundary conditions for our China study region. We use weekly flask and hourly surface observed CO_2 data provided by NOAA/ESRL, depending on data availability at each site. The ObsPack data product includes NOAA flask measurements at two Chinese sites (Extended Data Figure 1): Mt. Waliguan (WLG) and Shangdianzi (SDZ). The China Meteorological Administration (CMA) collect collocated CO_2 flask measurements at WLG and SDZ. WLG is located in the northeastern part of the Tibetan Plateau in western China. The station is situated far from industrial and populated centres, providing atmospheric information about emissions from the Eurasian continent (Zhou et al., 2004, 2005). The SDZ station is located 150 km northeast of Beijing (Fang et al., 2014) and provides a constraint on fluxes over the north China plain.

WDCGG network

We also use additional Chinese data at Hok Tsui (HKG) provided by Hong Kong Observatory (Fung et al, 2010) and accessed from the World Data Centre for Greenhouse Gases (WDCGG). HKG is situated at Hok Tsui Peninsular at the south eastern coast of Hong Kong Island (22°12'N, 114 °15'E, 60 m.a.s.l.), approximately 20 km from the urban centre of Hong Kong. There is no major road or settlement within 3 km of HKG, and therefore this site is considered to be representative of the larger-scale Pearl River Delta region.

JR-STATION network

We use CO_2 mole fraction data from the Japan-Russia Siberia Tall Tower Inland Observation Network (JR-STATION; Table S1, Extended Data Figure 1) over 2009-2016 (Sasakawa et al., 2010, 2013; Watai et al., 2010). We use these data to provide additional constraints to boreal Eurasia CO_2 fluxes.

The network was constructed by the Center for Global Environmental Research at the Japanese National Institute for Environmental Studies with the cooperation of the Russian Academy of Science. It includes eight towers over western Siberia and one in eastern Siberia at Yakutsk (Extended Data Figure 1). A nondispersive infrared (NDIR) analyser (LI-820, LI-COR, USA) is used to measure CO_2 mole fraction. These CO_2 observations data were calibrated against NIES 09 CO_2 scale.

For our experiments we use hourly data collected during 13:00-17:00 local time when we expect the atmosphere is well mixed and representative of a large geographical region (Saeki et al., 2013; Sasakawa et al., 2010).

Identifying Code	Location	Latitude	Longitude	Altitude (m)	Sampling height (m)
BRZ	Berezorechka	56.15° N	84.33° E	150	5,20,40,80
KRS	Karasevoe	58.25° N	82.42° E	50	35,67
IGR	Igrim	63.19° N	64.42° E	25	24,47
NOY	Noyabrsk	63.43° N	75.78° E	100	21,43
DEM	Demyanskoe	59.79° N	70.87° E	75	45,63
SVV	Savvushka	51.33° N	82.13° E	400	27,52
AZV	Azovo	54.71° N	73.03° E	100	29,50
VGN	Vaganovo	54.50° N	62.32° E	200	42,85
YAK	Yakutsk	62.09° N	129.36° E	130	11,70

Table S1 Atmospheric CO_2 sites over Siberia (JR-STATION) used in our calculations.

China Meteorological Administration *in situ* observation network over China

The China Meteorological Administration (CMA) is responsible for greenhouse gases observations under the framework of World Meteorological Organization/Global Atmosphere Watch programme (WMO/GAW).

We use hourly or weekly CO_2 data from 2009-2016 at seven stations in CMA, including one WMO/GAW global station, three WMO/GAW regional stations, and three CMA background stations. Location of the seven stations is illustrated in Extended Data Figure 1, Table S2.

Identifying Code	Location	Latitude	Longitude	Altitude (m)	Sampling height(m)	Representation area
WLG	Mt.Waliguan	36.12° N	100.06° E	3816.0	10, 80	Northeastern of the Tibetan Plateau
SDZ	Shangdianzi	40.65° N	117.12° E	293.3	10, 80	The North China Plain
LFS	Longfengshan	44.73° N	127.60° E	330.5	10, 80	Northeastern Plain
LAN	Lin'an	30.30° N	119.72° E	138.6	10, 50	Yangtze River Delta Economic Zone
SL	Shangri-La	28.00° N	99.40° E	3580.0	5*, 10, 50	southwest of China
JS	Jinsha	29.63 ° N	114.22 ° E	750.0	5*	Jianghan plain, Centre of China
AKDL	Akedala	47.10° N	87.93° E	562.0	5*	Junggar Basin, northwest of China

Table S2 Atmospheric CO_2 sites over mainland China used in our calculations. Sampling heights marked by an asterisk correspond to flask samples.

The Waliguan Baseline Observatory is the only WMO/GAW global atmospheric background station in China and also the only global atmospheric background station in inland Eurasia. The observatory is built on the top of mountain Waliguan in the northeast of the Tibet plateau. Greenhouse gases measurements from this site provide essential information on sources and sinks from within the Eurasian continent because of its unique location. The observed data at the site is also submitted to World Data Centre of Greenhouse Gases (WDCGG) and used for many scientific products (e.g., Global View database, WMO/GAW GHGs bulletin).

The LAN station is located at the centre of the Yangtze Delta area, 50 km from Hangzhou (the capital of Zheijang province) and 150 km from Shanghai. The Yangtze Delta economic zone represents one of the largest economic centres in China. The LFS station is in Northeast China, 175 km Southeast of Harbin City (the capital of Heilongjiang province) and provides a constraint on fluxes originating from the Northeastern Plain. The SDZ station is located 150 km northeast of Beijing (the capital of China) and provides a constraint on fluxes from the north China plain. The SL station is situated in Southwest China, 30 km north of Shangri-La county and 460 km from Kunming (the capital of Yunnan province). It resides in the transition zone of the Yunnan-Guizhou Plateau and the Tibetan Plateau. The JS station is located in the Jianghan plain located in the centre of China. AKDL is in the Xinjiang autonomous region in Northwest China and provides a constraint on the Gobi wetlands that reside between grassland and sparsely vegetated regions.

Instruments With the exception of JS and AKDL, all stations continuously measure CO_2 mole fractions using cavity ring-down spectroscopy (CRDS) analysers. The model of instruments was initially G1301 (Picarro Inc. USA), which we upgraded in 2015 to G2401 (Picarro Inc. USA).

Picarro CRDS instruments are commonly used by the global GHG measurement community because they have been proven suitable for making precise measurement of CO_2 mole fractions. The analysers have a highly linear response and are very stable (Crosson, 2008).

Figure S1 shows a schematic of the analytical system at Chinese stations. Ambient air is delivered to the instrument by a vacuum pump (N022, KNF Neuberger, Germany) via a dedicated 10 mm o.d. sampling line (Synflex 1300 tubing, Eaton, USA) and via a polytetrafluoroethylene (PTFE) 7 μm filter. The filtered ambient air is then pressurized to 1 atm and dried to a dew point of approximately - 60 °C by passing it through a glass trap submerged in a -70 °C ethanol bath (MC480D1, SP Industries, USA). An automated sampling module equipped with a VICI 8 port multi-position valve has been designed to sample from separate gas streams (standard gas cylinders, described below, and filtered ambient air). The residence time of the air from the top of the inlet to the analysers is less than 60 s. Further details about the measurement strategy are described by Fang et al. (2014).

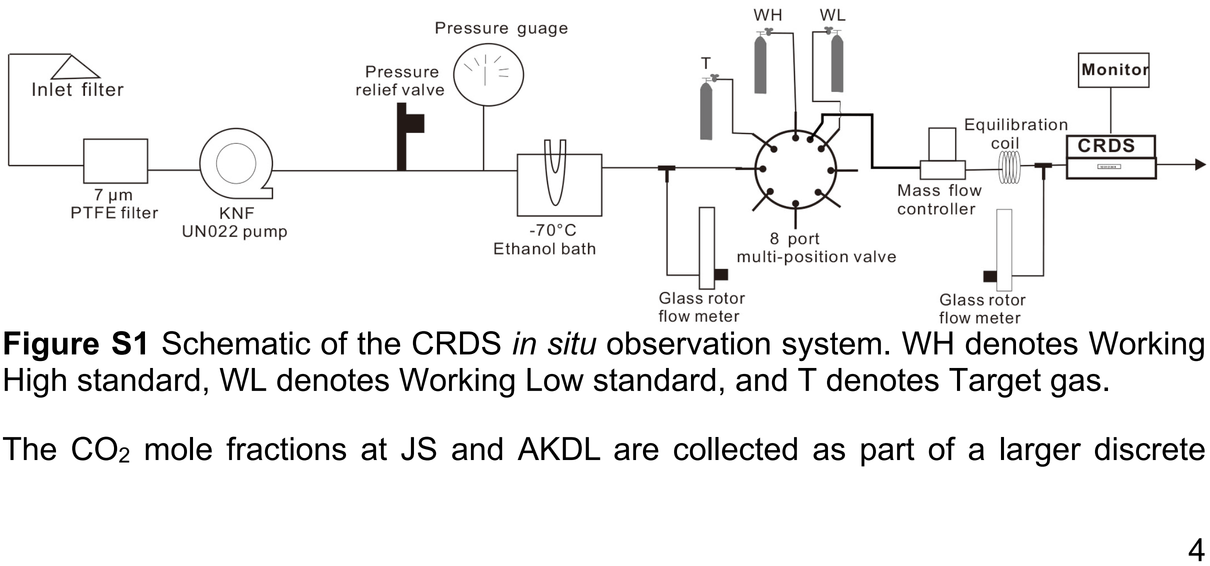

Figure S1 Schematic of the CRDS *in situ* observation system. WH denotes Working High standard, WL denotes Working Low standard, and T denotes Target gas.

The CO_2 mole fractions at JS and AKDL are collected as part of a larger discrete flask sampling programme in China (with eight stations), similar to the NOAA flask program (Dlugokencky et al., 1994). Two samples are collected in series using glass flasks and a portable battery powered sampling apparatus with a 5 m (a.g.l.) intake height. Weekly flask samples are collected at JS and AKDL and shipped to Beijing where they are analysed for CO_2. The CO_2 in flask samples are analysed by a Picarro G2301 (Picarro Inc. USA) system with a flow rate of 150 mL min^{-1}. The repeatability of flask-Picarro system is approximately 0.1 ppm.

Calibration The *in situ* CO_2 observation systems are routinely calibrated against the standards propagated by the WMO/GAW CO_2 Central Calibration Laboratory operated by NOAA/ESRL using the WMO X2007 scale (Zhao et al., 1997; Zhao and Tans, 2006). Carbon dioxide mole fractions are referenced to a Working High (WH) and a Working Low standard (WL) by using a linear two-point fit through the most recent standard gas measurements (WH & WL). Additionally, a calibrated cylinder filled with compressed ambient air is used as a target gas (T) to check the precision and stability of the system routinely.

All standard gases are pressurized in 29.5 L treated aluminium alloy cylinders (Scott-Marrin Inc) fitted with high-purity, two-stage gas regulators. An automated sampling module equipped with a VICI 8 port multi-position valve is designed to sample from separate gas streams (standard tanks and ambient air) at each of the station. The three standards are analysed by the system every 6 to 12 hours. Similarly, the flask measurements in Beijing lab are also linked to the WMO/GAW standards to guarantee the comparability.

Quality assurance and quality control All the measurements used in this study are provided under the framework of WMO/GAW Programme. To check the system performance at each station a calibrated cylinder filled with compressed ambient air is used as a target gas (T) to check the precision and stability of the system routinely. The bias between the assigned and measured value is evaluated and accepted when it is less than ±0.2 ppm. Otherwise, the ambient measurements are flagged as invalid, and no longer considered. Figure S2 shows examples of the bias at four of our stations. For the flask measurements analysed in our Beijing laboratory, we apply similar strategy to check the analytical quality.

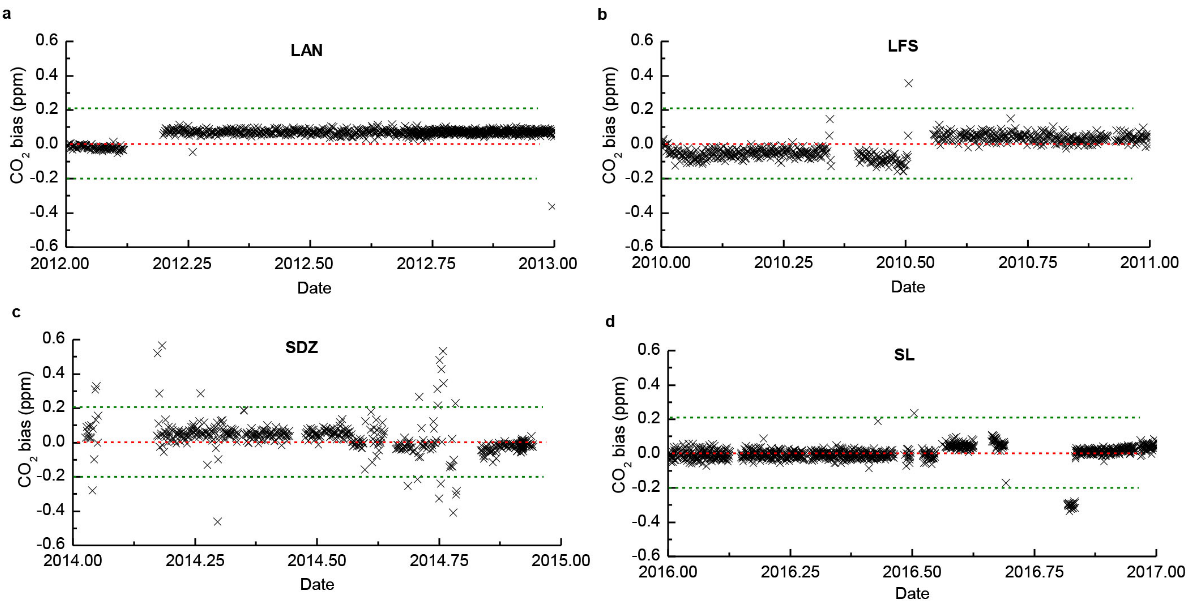

Figure S2 Typical examples of measurement bias relative to target measurements. Bias is described here as the difference between the assigned and the measured values for the target gas cylinders.

As a member of WMO/GAW programme we are obligated to participate in all comparison/audit activities sponsored by WMO/GAW to guarantee that our measurements continue to meet the goals of WMO/GAW. For example, results from the latest GAW round-robin (https://www.esrl.noaa.gov/gmd/ccgg/wmorr/) and the GAW World Calibration Centre (WCC) audit (https://www.empa.ch/documents/56101/250799/LAN_MOC_2016.pdf/e6d6ed18-f318-47f0-a6d2-60f272bf9845) indicate that the *in situ* measurements used in this study meet the goals of WMO/GAW. Figure S3 shows an example of the CO_2 measurements audited at the LAN station, with bias reported between our measurements and the GAW WCC standards for six travel standards.

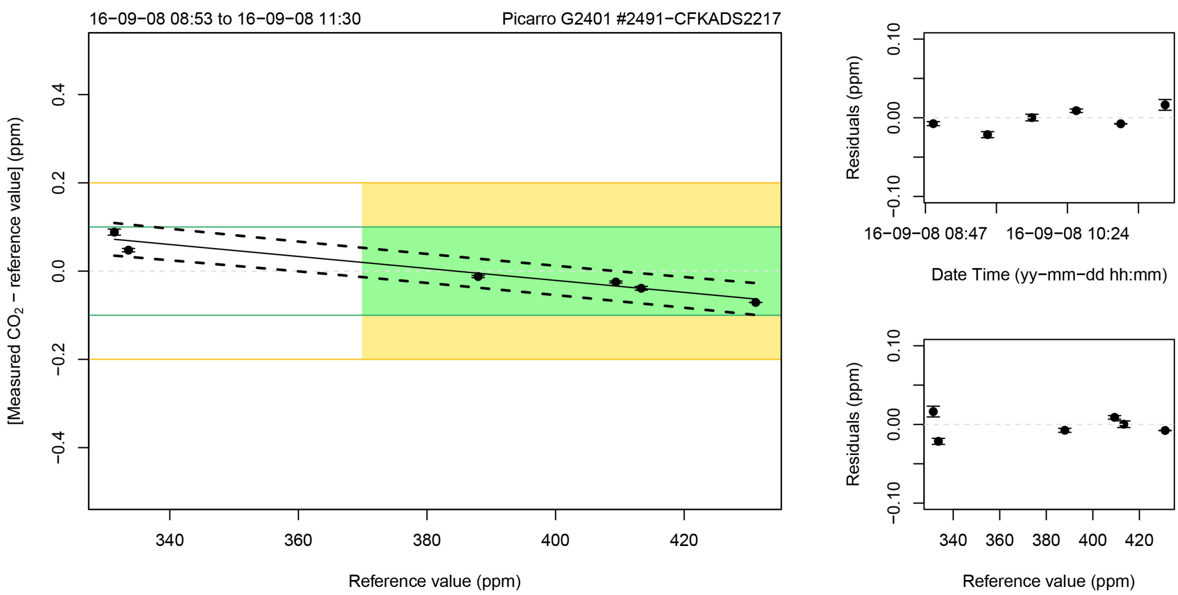

Figure S3 Audited CO_2 measurements at LAN station in China. Over the tested CO_2 mole fraction range, the measurements at LAN meet the ±0.1 ppm compatibility goals of WMO/GAW denoted by the green shaded region; the yellow shaded regions shows the extended goal. This figure is adapted with permission from the audit report of WMO/GAW WCC: https://www.empa.ch/documents/56101/250799/LAN_MOC_2016.pdf/e6d6ed18-f318-47f0-a6d2-60f272bf9845.

Data processing Because of the dead volume in the plumbing of the *in situ* system, the response of the system is not stable until one minute after switching to a different gas source using the multi-position valve. Acknowledging this, we sequentially connect every standard gas cylinder to the system for 5 minutes. Consequently, the raw ambient data are separated into 5-minute segments. The concentration routine discards the first 3 minutes of data and only uses the mean of the last 2 minutes to calculate the concentration through the latest linearity regression factors. Because the CRDS system measures every 2-3 seconds, the number of measurements in the last 2 minutes is sufficient to represent the conditions in identical 5 minutes segments. Invalid data caused by instrumental malfunction are checked with information from the station logbook. After computing CO_2 mole fractions, the data are manually inspected. Occasionally, information in the station logbook identifies periods when measurements are affected by analytical or sampling problems including poor instrument performance or local influences such as nearby fires, vehicles, and cattle. After these steps, all of the 5-minute data are combined into hourly averages that we use in our study.

Data filtering The CO_2 observations at the sites are unavoidably influenced by local sources and sinks. Identification of regionally-representative CO_2 measurements that are minimally influenced by local anthropogenic emissions or land biosphere is therefore essential. To distinguish the CO_2 observations influenced by anthropogenic emissions adjacent to a station (villages, traffic emissions, etc.) and local vegetation canopy (field, forest, etc), as used in previous studies (Fang et al., 2014, 2015), we filter the hourly CO_2 observations by considering the meteorological factors, local topography, and anthropogenic activities.

We employ a three-stage data filtering process. First, we only use data during daylight hours, e.g. 10:00 to 16:00 (local time) at LAN and from 09:00 to 16:00 (local time) at LFS and SDZ, which have better representative conditions. Measurements of CO_2 at LAN station from two levels (10 and 50 m a. g. l.) have shown that the vertical difference between the two levels is minimum (less than 0.2±0.2 ppm) between 10:00 and 16:00 and is stable. Thus, the data during this period of the day are selected when the boundary layer is high and the vertical mixing is strong (e.g., Figure S4 for LAN and LFS stations). For other stations, we apply a similar method.

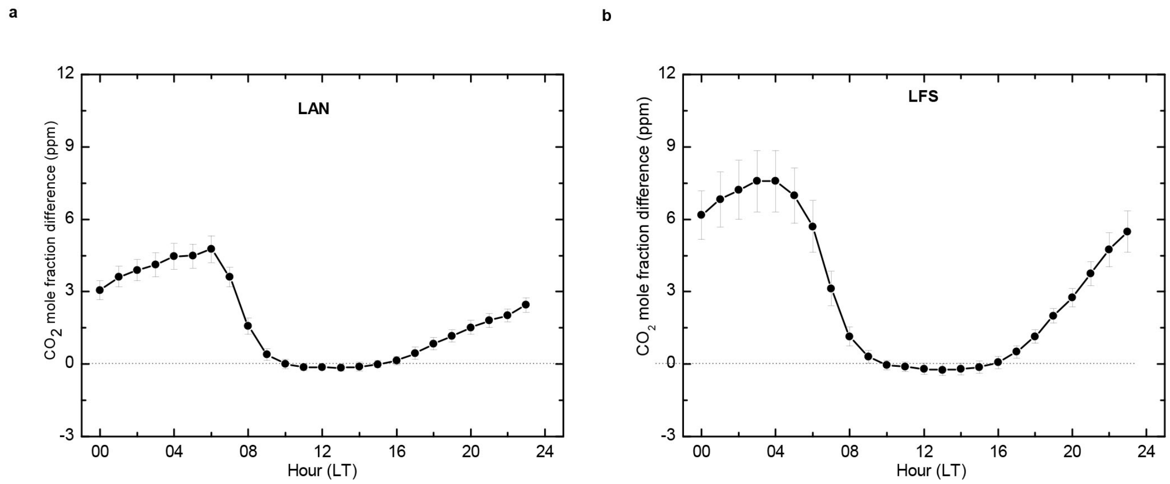

Figure S4 Example of diurnal differences of hourly CO_2 mole fractions between the 10 m and the top of each tower at **a** LAN and **b** LFS station. Error bars denote confidence intervals of 95%.

Second, guided by nearby potential contamination sources, CO_2 observations are determined to be locally representative when surface winds prevail from adjacent local sources/sinks such as villages, factories, and paddy rice fields. The rejected wind sectors (local representative) are flagged in four seasons at all of the stations. For example, the data when local surface winds are from SSW-SW and N sectors are flagged at LAN station to exclude the transports from the Lin'an town located at 6 km southwest and the small factory (north of the station, 1.4 km away), where charcoal is manufactured from burning bamboo wood.

Finally, to minimize the influence of very local sources and sinks, the remaining data is filtered by discarding data when the local surface wind speed is lower than 1.5 m s^{-1} (Beaufort scale 0 and 1). For the WLG station, we also flag the CO_2 records when the wind speed is higher than scale 6 (>10.8 ms^{-1}), because the higher wind speed may also transport emissions from much broader regions (e.g., Xining and Lanzhou) at WLG. After our three-step filter, the resulting hourly data is considered to be regionally representative and are used in our study.

For the discrete flask samples at JS and AKDL, anomalous data caused by improper sample collection operations (Lang et al., 1990) or error in analysis (flagged by QC), are discarded. When the difference between pair samples is larger than 0.4 ppm, corresponding to three times the mean pair agreement, we consider one or both flasks are contaminated and discard the measurement. After this processing, the means of flask sample pairs are used in this study.

Extended Data Figure 1 shows the hourly/weekly mean observations from the sites we use across China. We use some flask observations to fill gaps left by the CRDS system in SL.

GOSAT and OCO-2 Satellite Observations

We use column CO_2 data collected by the Japanese Greenhouse gases Observing SATellite (GOSAT) and the NASA Orbiting Carbon Observatory (OCO-2). GOSAT was launched in January 2009 in a 666-km altitude sun-synchronous orbit with a local overpass time over China around 13:00 in its descending node (Kuze et al, 2016). OCO-2 was launched on July 2, 2014 and inserted into the front of the 705-km Afternoon Constellation (A-Train) on August 3, 2014. It has a mean local overpass time of 13:36 in its ascending node and has a 16-day (233-orbit) ground-track repeat cycle (Crisp, 2015). Recent work has shown that radiance measurements from earlier versions of these independent datasets (ACOS-GOSAT B7.3 and ACOS-OCO2 B7) are generally within 5% for all bands when they view the same scene at the same time (Kataoka et al, 2017).

The GOSAT Thermal And Near-infrared Sensor for carbon Observation (TANSO) comprises two instruments: a Fourier Transform Spectrometer (FTS) and the Cloud and Aerosol Imager. The FTS observes five across-track nadir pixels (three across-track pixels after August 2010) with a footprint diameter of 10.5 km, an across-track spacing of about 100 km, and an along-track spacing of 90–280 km. This results in an approximate three-day revisit time. We use the column-averaged dry air mole fraction of atmospheric CO_2(X_{CO2}) full-physics retrievals from v7.3 data (downloaded from GES DISC/GOSAT) product from the Atmospheric CO_2 Observations from Space (ACOS) from the Jet Propulsion Laboratory (JPL, O'Dell et al., 2012).

OCO-2 comprises three imaging grating spectrometers for three bands: two shortwave infrared CO_2 bands and an O_2 band (Crisp et al.,2008). The OCO-2 employs three observation modes: nadir, sun glint (specular reflection), and target. For routine science operations, OCO-2 either points the instrument boresight to the local nadir or towards sun glint. The spatial resolution of individual measurements is < 3km^2 (Eldering et al., 2017). We use ACOS v8r OCO-2 X_{CO2} data (O'Dell et al.,2018) for this work (downloaded from GES DISC/OCO-2).

Extended Data Figure 4 shows the spatial coverage of clear-sky X_{CO2} data observed by GOSAT over China during December 2014 to November 2015. It shows a seasonal dependence, mainly reflecting changes in solar zenith angle and cloud coverage. There are very few data in winter over the northern part of China. Extended Data Figure 4 also shows that for the same period there are many more clear-sky observations from OCO-2, reflecting the smaller ground footprint of OCO-2 measurement.

2 Atmospheric Transport Model and ensemble Kalman Filter

We use the GEOS-Chem global chemistry transport model (v9.02, http://acmg.seas.harvard.edu/geos/index.html) to relate *a priori* surface fluxes to atmospheric CO_2 observations. We run the model at a spatial resolution of 4^o latitude by 5^o longitude and 47 vertical levels, driven by GEOS-5 (2009-2012) and GEOS-FP (2013-2016) meteorological analyses from the Global Modeling and Assimilation Office at NASA Goddard Space Flight Centre. We use *a priori* flux inventories to describe: monthly fossil fuel emissions, (Oda and Maksyutov, 2011); weekly biomass burning emissions (van der Werf et al., 2010); climatological monthly oceanic surface fluxes (Takahashi et al., 2009), and three-hourly terrestrial biosphere-atmosphere fluxes (Olsen and Randerson, 2004).

We use an ensemble Kalman filter (Feng et al., 2009, 2011, 2017) to infer *a posteriori* surface CO_2 fluxes from atmospheric CO_2 data. We estimate monthly fluxes on a spatial distribution based on TransCom-3 (Gurney et al., 2002) with each continental and ocean region further divided into sub-regions. On the global scale, we estimate fluxes for 475 land regions and 317 ocean regions (totally 792 regions, Figure S5) for each month between January 2009 and December 2016, resulting in a state vector length of 76,032. Over China, we infer fluxes at the spatial resolution of the atmospheric transport model, corresponding to 58 geographical regions. We discuss below the role of flux error correlations between neighbouring grid points.

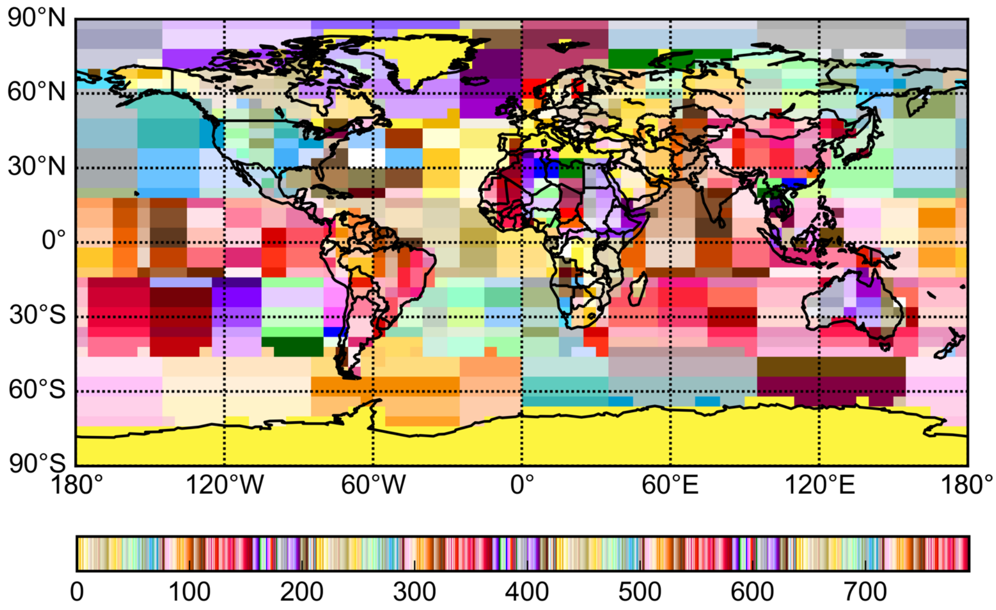

Figure S5 A map of the 792 geographical regions used in our inversions.

Here, we outline the approach we use to estimate annual uncertainty estimates from the ensemble Kalman filter with a lagged window. This approach provides a simple approximation for estimating the uncertainty in multi-year *a posteriori* flux estimates. It is based on the assumption that direct temporal error correlations are introduced by sharing the same observation constraints, and hence they are relatively small when the time gap is larger than the lag window. This approach retains the correlations within our 4-month lag window while propagating indirect correlations step-by-step through overlaid months of the moving lag window.

Following the sequential data assimilation theorem, observations for the whole experiment period are assimilated sequentially monthly. *A posteriori* estimates from each step are used as the *a priori* to assimilate observations in the following month. By assimilating observations at month *i*, we obtain the *a posteriori* error covariance $\mathbf{P}_i^a$:

$$\mathbf{P}_i^a = (\mathbf{I} - \mathbf{K}_i\mathbf{H}_i)\mathbf{P}_i^f, \quad (1)$$

where $\mathbf{H}_i$, a subset of the full Jacobian $\mathbf{H}$, projects flux estimates corresponding to month *i* to observation space, and the Kalman gain matrix $\mathbf{K}_i$ for step *i* is determined by

$$\mathbf{K}_i = \mathbf{P}_i^f \mathbf{H}_i^T [\mathbf{H}_i \mathbf{P}_i^f \mathbf{H}_i^T + \mathbf{R}_i]^{-1}, \tag{2}$$

where $\mathbf{R}_i$ represents the observation error covariance subset for month *I*. $\mathbf{P}_i^f$ is the *a priori* flux for step *i*, which is the *a posteriori* error covariance from step *i-1*. We have assumed individual observations are independent.

We now introduce flux perturbation matrices, $\Delta\mathbf{X}_i^f$ and $\Delta\mathbf{X}_i^a$, as the square root of *a priori* and *a posteriori* error covariances, respectively: $\mathbf{P}_i^f = \Delta\mathbf{X}_i^f(\Delta\mathbf{X}_i^f)^T$, and $\mathbf{P}_i^a = \Delta\mathbf{X}_i^a(\Delta\mathbf{X}_i^a)^T$. As a result, we can re-write Eq. 2 as:

$$\Delta\mathbf{X}_i^a(\Delta\mathbf{X}_i^a)^T = \Delta\mathbf{X}_i^f[\mathbf{I} - \Delta\mathbf{Y}_i^T(\Delta\mathbf{Y}_i^T\Delta\mathbf{Y}_i + \mathbf{R}_i)^{-1}\Delta\mathbf{Y}_i](\Delta\mathbf{X}_i^f)^T, \tag{3}$$

where $\Delta\mathbf{Y}_i = \mathbf{H}_i\Delta\mathbf{X}_i^f$ represents the spread of the model concentration values as the response to flux perturbation matrix $\Delta\mathbf{X}_i^f$. Following the Ensemble transform Kalman filter approach (Bishop et al., 2001), we introduce a transform matrix $\boldsymbol{\alpha}_i$:

$$\boldsymbol{\alpha}_i(\boldsymbol{\alpha}_i)^T = [\mathbf{I} - \Delta\mathbf{Y}_i^T(\Delta\mathbf{Y}_i^T\Delta\mathbf{Y}_i + \mathbf{R}_i)^{-1}\Delta\mathbf{Y}_i], \tag{4}$$

allowing us to rewrite Eq. 3 as:

$$\Delta\mathbf{X}_i^a(\Delta\mathbf{X}_i^a)^T = (\Delta\mathbf{X}_i^f\boldsymbol{\alpha}_i)(\Delta\mathbf{X}_i^f\boldsymbol{\alpha}_i)^T = \Delta\mathbf{X}_{i+1}^f(\Delta\mathbf{X}_{i+1}^f)^T, \tag{5a}$$

or

$$\Delta\mathbf{X}_{i+1}^f = \Delta\mathbf{X}_i^a = \Delta\mathbf{X}_i^f\boldsymbol{\alpha}_i\ . \tag{5b}$$

As shown in Eqs. 4 and 5, the transform matrix, $\boldsymbol{\alpha}_i$ mainly reduces the spread of the flux perturbation matrix due to the assimilation of observations at month *i*. However, $\boldsymbol{\alpha}_i$ is usually non-diagonal, with off-diagonal values representing contributions to $\Delta\mathbf{Y}_i$ from flux perturbations at different time and locations even though we assume $\mathbf{R}_i$ is diagonal.

After repeating the above steps to assimilate observations through the whole experiment period of *m* months, we obtain the final *a posteriori* error covariance:

$$\mathbf{P}^a = \Delta\mathbf{X}_m^a(\Delta\mathbf{X}_m^a)^T = (\Delta\mathbf{X}_0\boldsymbol{\alpha}_t)(\Delta\mathbf{X}_0\boldsymbol{\alpha}_t)^T, \tag{6}$$

where the final transform matrix $\boldsymbol{\alpha_t}$ is calculated iteratively by propagating from month *i*=1 to m:

$$\boldsymbol{\alpha_t} = \prod_{\boldsymbol{i=1}}^{m}\boldsymbol{\alpha}_i. \tag{7}$$

With the exception of our assumption of independent observations, no explicit approximation has been introduced in Eqs. 6 and 7 for determining the full *a posteriori* error covariance. However, we have a total of 76032 (8 years x 12 months x 792 regions) variables. Any direct numerical computation using Eqs.6 and 7 would be very expensive, partially because the projection of the flux perturbations to the observation space (i.e., $\Delta\mathbf{Y}_i \sim \mathbf{H}_i \Delta\mathbf{X}_i^f$) also involves atmospheric modelling of tracer transport using a global 3D chemistry transport model.

In our study, we use a moving lag window of 4 months to reduce computational cost. This is a common approach that assumes that observations at one time provide only weak constraints on past fluxes on time periods longer than 4 months, i.e. contributions are too dilute. It reduces the effective variable number to 3168 (4 months x 792 regions) within each assimilation window, so that the corresponding transform matrix $\boldsymbol{\alpha}_i^w$ has a much reduced dimension of 3168×3168, even when a full representation of the perturbation matrix is used (Feng et al., 2009, 2016). Further, the windowed transform matrix $\boldsymbol{\alpha}_i^w$ is one of the step-by-step outputs explicitly estimated in our current study based on an ensemble approach (Feng et al., 2009, 2016).

During post-processing, we use Eq. 7 to propagate the individual transform matrices $\boldsymbol{\alpha}_i^w$ in to the final *a posteriori* error covariance (Eq. 6). Constraint by observations on flux estimates outside their lag window is relatively weak and we ignore those contributions. As such, we can approximate the full-size $\boldsymbol{\alpha}_i$, which describes the impacts on the full flux estimate uncertainty by observations at month *i*, by assuming:

$$\boldsymbol{\alpha}_i \approx \begin{bmatrix} \mathbf{I}_{1,i-4} & \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \boldsymbol{\alpha}_i^w & \mathbf{0} \\ \mathbf{0} & \mathbf{0} & \mathbf{I}_{i+1,n} \end{bmatrix}, \tag{8}$$

where $\mathbf{I}_{1,i-3}$, and $\mathbf{I}_{i+1,n}$ are identity matrices for preceding months 1 to *i*-4, and for the subsequent months *i*+1 to *n*, respectively. By applying the (approximated) full size $\boldsymbol{\alpha}_i$ for each step to Eq. 7, we are able to construct the final transform matrix $\boldsymbol{\alpha}_t$. Together with the assumed *a priori* flux perturbation $\Delta\mathbf{X}_0$, we can then estimate the full *a posteriori* error covariance via Eq. 6, enabling us to calculate aggregated uncertainty for selected region and time period.

Our reported uncertainty for annual mean regional flux estimates should be considered as underestimates. Similar to alternative methods of estimating *a posteriori* errors, we are subject to the accuracy of often poorly quantified errors that describe the *a priori* and the atmospheric transport model.

3 Results

The impact of *in situ* data on understanding Chinese CO_2 fluxes

We use a simple error metric to determine the improvement in our understanding of Chinese CO_2 fluxes from using the additional data over Siberia and China: $\gamma = 1 - \sigma^a/\sigma^f$ (Palmer et al, 2000), where σ^a and σ^f denote the *a posteriori* and *a priori* variance uncertainties, respectively.

Figure S6 shows that the γ metric from the NOAA/ESRL and the additional reduction from using the JR-STATION and Chinese data. We find that the Siberian data improve CO_2 fluxes over most of boreal Eurasia, including the geographical region immediately upwind of China. This allows the new Chinese data to constrain flux estimates across China, as we report in detail below.

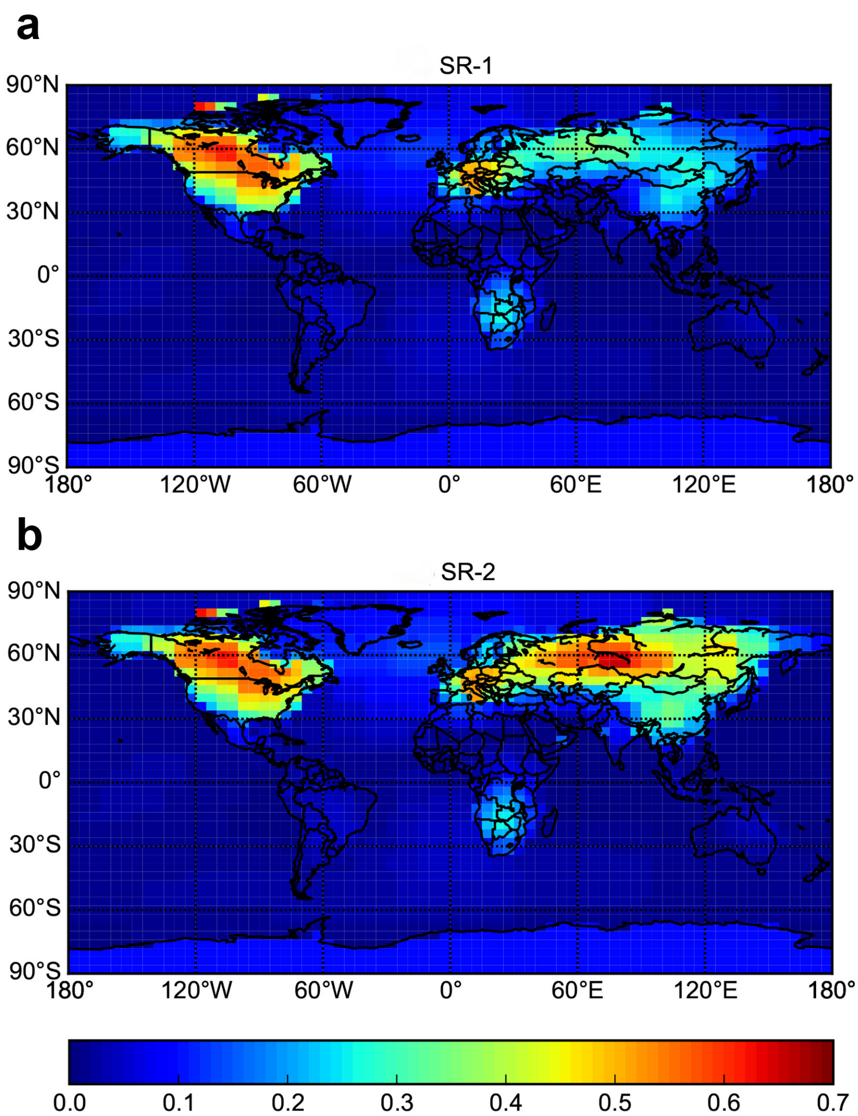

Figure S6 Example error reduction (γ metric) for April 2011. Top panel shows the γ metric after ingesting NOAA/ESRL ObsPack data (inversion SR-1, see main paper), and the bottom panel shows the γ metric after ingesting ObsPack + JR-STATION + WDCGG and CMA data (inversion SR-2). Higher γ values denote a larger reduction in flux uncertainties.

<u>Seasonal difference between SR-1 and SR-2 inversions</u>

Figure 1 shows that inversion SR-1 generally underestimates the annual uptake in Southwest China and the emission over eastern and northern China (north of 30°N, east of 110°E).

Figure S7 shows that the seasonal SR-2 minus SR-1 differences averaged over 2010-2016. We find that the SR-1 underestimates uptake in Southwest China throughout the year, and underestimates uptake in Northeast China mainly during summer months. This is further discussed below when we report changes in leaf phenology. We also find that SR-1 underestimates net emissions over eastern and northern China (38-54°N, 115-135°E) during spring, autumn, and winter months.

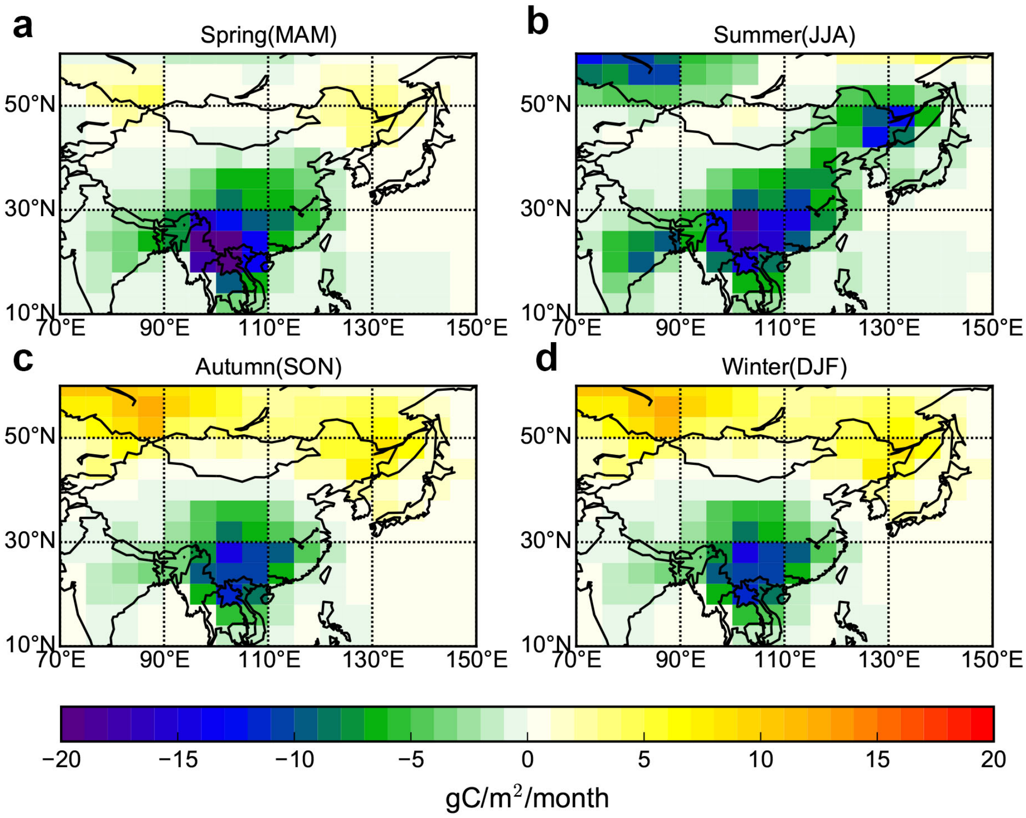

Figure S7 Seasonal mean differences of SR-2 minus SR-1 inversions during 2010-2016 (gC/m^2/month).

<u>Sensitivity of land biosphere fluxes to assumed *a priori* anthropogenic emissions</u>

Our inverse model calculations estimate the net CO_2 flux. We determine the land biosphere flux by subtracting emission estimates from fossil fuel combustion and cement production. This is a common approach (Peylin et al., 2013; Thompson et al.,2016; Jiang et al.,2016), based on an assumption about our knowledge of fossil fuel emissions. Consequently, our biosphere fluxes depend on what we assume for anthropogenic emissions.

Here, we report a range of China fossil fuel flux estimates using inventories from the Carbon Dioxide Information Analysis Center (CDIAC, Boden et al., 2017); Emissions Database for Global Atmospheric Research (EDGAR V4.3.2, Janssens-Maenhout et al., 2017); International Energy Agency (IEA, 2017); Liu et al., (2015); Shan et al., (2018); and the Open-Data Inventory for Anthropogenic Carbon dioxide (ODIAC, 2016). The data published by CDIAC, EDGAR and ODIAC are similar and larger than the data from IEA, 2017 and Liu et al (2015) (Table S3). We scale ODIAC emissions over China by 88% for our control run, guided by recent Chinese sectoral emission estimates from Shan et al., (2018). We retain the seasonal and spatial distributions of ODIAC emissions over China.

	CDIAC	EDGAR	IEA	Liu	Shan	ODIAC	88% ODIAC
2010	2.48	2.52	2.25	2.11	2.16	2.49	2.19
2011	2.74	2.75	2.48	2.28	2.39	2.75	2.42
2012	2.82	2.81	2.52	2.43	2.48	2.82	2.48
2013	2.88	2.93	2.63	2.50	2.60	2.92	2.57
2014	2.90	2.97	2.64		2.58	2.93	2.58
2015	2.87	2.94	2.64		2.53	2.92	2.57
2016	2.87	2.93				2.92	2.57

Table S3 Annual Chinese estimates of fossil fuel and cement production (PgC/yr) from 2010 to 2016. IEA values include only fossil fuel emissions.

A priori annual fossil fuel emissions for China, taken from the ODIAC inventory, represent 2.49-2.93 PgC/yr. As described above, for our control run we use 88% of the ODIAC emission estimates, so the fossil fuel emissions are 2.19-2.58 PgC/yr. Including fluxes from the ocean, biomass burning, and the land biosphere (section 2) results in a net *a priori* flux of 2.33-2.61 PgC/yr.

Results and comparison with previous studies

Table S4 shows our annual mean *a posteriori* land biosphere CO_2 uptake for SR-1 and SR-2 are -0.66±0.52 PgC/yr and -1.11±0.38 PgC/yr, respectively, during 2010-2016. In comparison, annual mean *a posteriori* land biosphere CO_2 uptake estimates inferred from GOSAT-ACOS and OCO2 are -0.83±0.47 PgC/yr and -0.88±0.43 PgC/yr during 2010-2015 and 2015-2016, respectively, crudely separating year to account for the 2015/2016 El Niño.

As noted in the main paper (Table 1), our SR-2 estimate for the land biosphere uptake of CO_2 inferred from the new CMA *in situ* data (section 1) is higher than previous estimates. Piao et al (2009) used biomass and soil carbon inventories extrapolated by satellite observations of leaf phenology, process-based ecosystem models, and atmospheric inversions to infer independent CO_2 flux estimates that range from -0.19 to -0.26 PgC/yr for Chinese terrestrial ecosystems over the 1980s and 1990s. They attributed 65% of this carbon sink to southern China due to regional changes in climate, large-scale reforestation and shrub recovery. Tian et al (2011) used a process-based ecosystem model driven by meteorological and atmospheric composition data and by knowledge of regional land use and management practices. They estimated a Chinese CO_2 sink of -0.18 PgC/yr with a 95% confidence range from -0.06 PgC/yr to -0.26 PgC/yr. Wang et al (2015) used climate data, spatial distributions of ecosystems, and social statistics to infer a mean Chinese terrestrial biosphere carbon sink of -0.97 PgC/yr from 2001 to 2010. Zhang et al., (2014) and Jiang et al (2016) inferred the Chinese terrestrial biosphere CO_2 sink using atmospheric data with four sites over China (WLG, LAN, LFS, SDZ). Zhang et al., (2014) estimated a mean sink of -0.33 PgC/yr over the period 2001-2010 and Jiang et al. (2016) estimated a mean sink of -0.45 PgC/yr over the period 2006-2009, ranging from -0.39 to -0.51 PgC/yr.

Other prominent studies on this topic have reported CO_2 flux estimates for East Asia, which include CO_2 terrestrial biosphere fluxes from China aggregated with those from Japan, North and South Korea and Mongolia. Piao et al (2012) used similar approaches to those used by Piao et al (2009) but with a larger ensemble of estimates to study a longer period from 1990 to 2009. They estimated the terrestrial carbon sink for East Asia to be -0.293±0.033 PgC/yr (inventory based approach), -0.224±0.141 PgC/yr (process-based ecosystem models), and -0.270±0.507 PgC/yr (inferred from atmospheric data). Thompson et al. (2016) used an ensemble of terrestrial carbon flux estimates from several inversions to calculate a mean Asian land sink of -0.46 PgC/yr, ranging from -0.70 PgC/yr to -0.24 PgC/yr, for the period 1996-2012. They noted this sink was mostly located in East Asia where the mean CO_2 sink increased by 0.56 PgC/yr from 1996-2001 to 2008-2012.

Our results for the standard inversion SR-1 are within the range of those reported by Thompson et al (2016), as expected since they use similar data. The additional CMA data provide strong constraints on Southwest China (SR-2) where other studies have noted there is strong carbon uptake but have not had sufficient data to quantify those regional fluxes.

	A priori				SR-1	SR-2	GOSAT-ACOS	GOSAT ACOS +SR-2	OCO2-ACOS
	Total	BS	FF	$1-\sigma$	BS	BS	BS	BS	BS
2010	2.23	0.04	2.19	0.97	-0.60±0.51	-0.99±0.38	-0.60±0.46	-0.95±0.32	-
2011	2.33	-0.11	2.42	1.01	-0.74±0.53	-1.32±0.36	-0.93±0.49	-1.30±0.32	-
2012	2.49	0.01	2.48	1.01	-0.74±0.53	-1.25±0.39	-0.69±0.51	-1.12±0.34	-
2013	2.59	0.02	2.57	1.02	-0.61±0.55	-0.87±0.45	-0.88±0.47	-0.96±0.36	-
2014	2.61	0.03	2.58	0.99	-0.68±0.49	-1.08±0.39	-0.93±0.48	-1.09±0.33	-
2015	2.54	-0.03	2.57	1.03	-0.52±0.54	-1.03±0.34	-0.92±0.44	-1.02±0.31	-0.85±0.45
2016	2.58	0.01	2.57	0.93	-0.77±0.47	-1.25±0.31	-	-	-0.91±0.41
Mean	2.48	0	2.48	0.99	-0.66±0.52	-1.11±0.38	-0.83±0.47	-1.07±0.33	-0.88±0.43

Table S4 Annual mean CO_2 fluxes (PgC/yr) inferred from *in situ* and remote sensing data. *A priori* values for the total denote the sum of values from the terrestrial biosphere (BS) and fossil fuel and cement manufacture (FF). The 1-σ value uncertainty corresponds to the total value. Inversion SR-1 denotes NOAA ObsPack *in situ* data (including one site over China); inversion SR-2 denotes *in situ* data from NOAA ObsPack, Siberia, and from China; GOSAT-ACOS denotes GOSAT XCO_2 data retrieved by the ACOS team; an OCO2-ACOS denotes OCO-2 XCO_2 data retrieved by the ACOS team.

Sub-continental Chinese CO_2 flux estimates

To report sub-continental CO_2 fluxes, we divide the Chinese mainland into eight geographical regions (Extended Data Figure 2), based on land use cover and economic regions. The eight sub-regions are Northwest China (NWC), North China (NC), Northeast China (NEC), Tibet Plateau (TP), Centre China (CC), East China (EC), Southwest China (SWC) and Southeast China (SEC).

Table S5 shows that *a posteriori* CO_2 fluxes from our defined sub-continental regions are weakly correlated with neighbouring regions, with the highest values generally with adjacent regions. We also give the correlation between one Siberia region (SIB,54°N-66°N, 70°E-85°E) and eight sub-continental regions in China. The Siberia region is weakly correlated with sub-continental regions in China.

	Region	NW	North	NE	TP	CC	East	SW	SE	SIB
	NW	+1.00								
	North	-0.062	+1.00							
	NE	-0.035	0.355	+1.00						
Jan	**TP**	0.218	0.069	-0.059	+1.00					
	CC	0.040	0.388	-0.060	0.534	+1.00				
	East	-0.048	0.669	0.069	0.120	0.564	+1.00			
	SW	-0.032	0.141	-0.052	0.401	0.528	0.413	+1.00		
	SE	-0.050	0.201	-0.028	0.063	0.370	0.493	0.837	+1.00	
	SIB	-0.014	-0.077	-0.121	-0.056	-0.036	-0.017	0.0	0.006	+1.00
	Region	**NW**	**North**	**NE**	**TP**	**CC**	**East**	**SW**	**SE**	**SIB**
	NW	+1.00								
	North	-0.049	+1.00							
	NE	-0.034	0.364	+1.00						
Jul	**TP**	0.230	0.117	-0.050	+1.00					
	CC	0.039	0.383	-0.058	0.551	+1.00				
	East	-0.042	0.654	0.065	0.179	0.568	+1.00			
	SW	-0.033	0.129	-0.062	0.314	0.604	0.387	+1.00		
	SE	-0.054	0.177	-0.048	0.089	0.383	0.501	0.652	+1.00	
	SIB	-0.014	-0.078	-0.070	-0.066	-0.043	-0.025	0.001	0.001	+1.00

Table S5 *A posteriori* CO_2 flux correlation coefficients of SR-2 between neighbouring regions during contrasting months, January and July 2011.

Figure S8 shows annual net CO_2 flux estimates for each sub-continental regions shown in Extended Data Figure 2. *A posteriori* uncertainties are generally much smaller than *a priori* values, as expected, with the smallest values for the SR-2 inversion that reflects the added information from the new sites.

We find that TP and SWC are the regions which have a continuous annual net CO_2 sink over the seven study years. The large net uptake in SWC corresponds to low fossil fuel emission and high biosphere uptake in this region (Extended Data Figure 2). The other six geographical regions represent annual CO_2 emissions. The largest emissions are from EC, NC, and SEC, consistent with the population distribution and economic zones: Yangtze River Delta, Circum-Bohai Sea, and the Pearl River Delta. EC accounts for approximately 52% of net Chinese CO_2 emissions, despite significant land biosphere uptake. EC, SWC, SEC, NEC and CC are important regions for biosphere uptake, with SWC representing the largest annual value.

Extended Data Figure 2 shows the monthly mean biosphere CO_2 fluxes. For many of the regions the SR-1 inversion tracks *a priori* values throughout the year. The SR-2 inversion generally shows larger variations throughout each season compared with the SR-1 inversion. Over SWC *a posteriori* CO_2 fluxes are negative for most of the year, leading to a large, persistent annual net CO_2 uptake over this region, accounting for about 32% of the biosphere uptake over whole China.

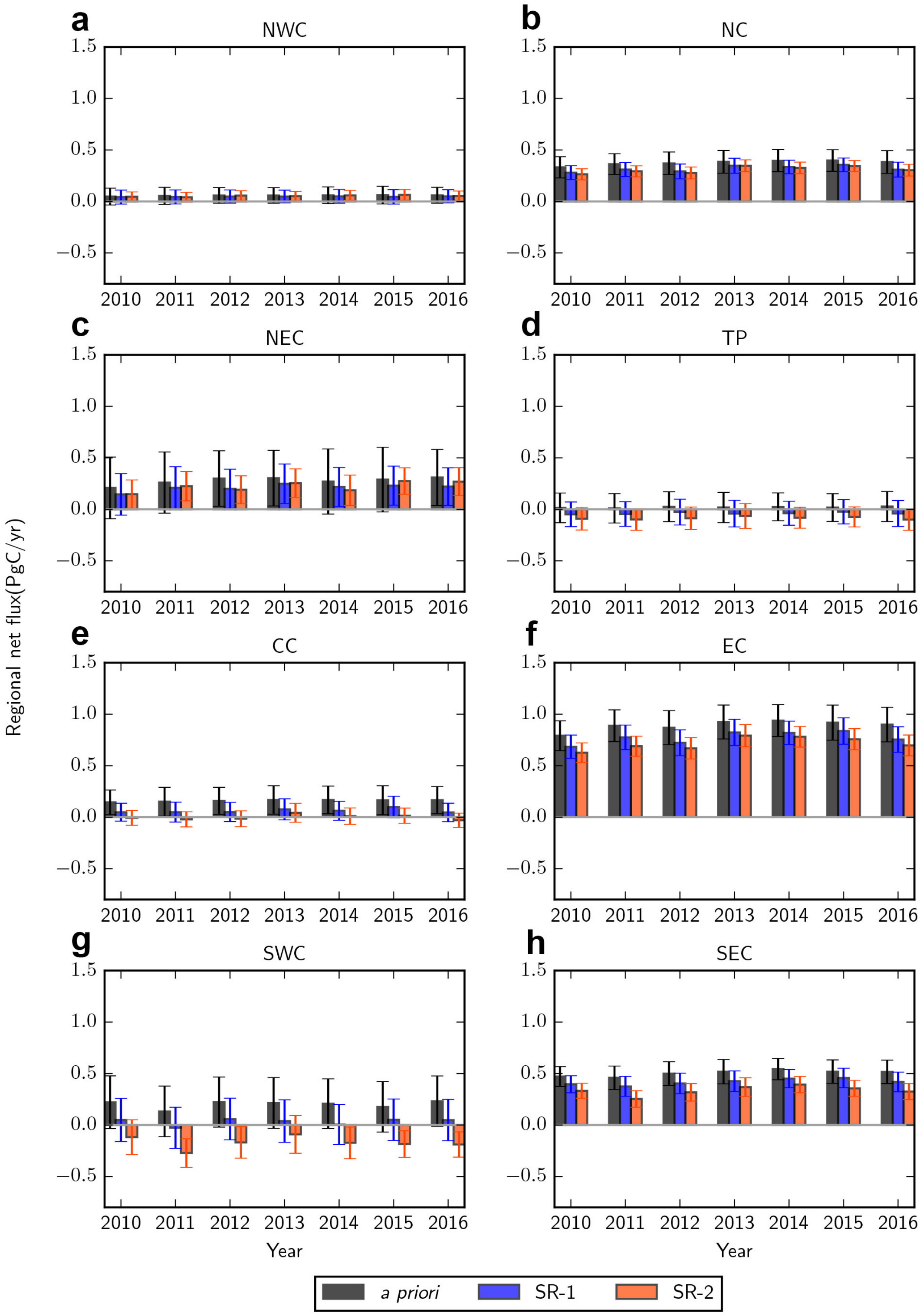

Figure S8 Annual regional *a priori* and *a posteriori* net CO_2 fluxes over China during 2010—2016. *A posteriori* fluxes are inferred from data used in SR-1 and SR-2 inversions.

<u>Sensitivity tests</u>

A posteriori values are reported as the mean annual estimate for period 2010-2016, inclusive.

In our assimilation of (continuous) *in situ* observations, we sample model surface CO_2 mixing ratios at the time (hour) and location of each observation and then optimally fit the resulting model daytime means with the mean observations between local time 09:00 to 16:00. Uncertainty of the mean observation is estimated by taking in account both the instrument (random) error, and the representation error that is mainly due to coarse model resolutions:

$\varepsilon_t^2 = \varepsilon_{obs}^2 + \sigma_m^2$,

where ε_{obs} is the instrument (random) error of the observation daytime mean, and σ_m represents the standard deviation between model and observed CO_2 mole fractions between 09:00 and 16:00 local time. As a result, less weight is given to observations for which (comparatively coarse) model simulations are unable to reproduce transient and small-scale variations observed by the *in situ* sites.

To further remove observations with variations that are not described by the model, we discard observations when there are fewer than three (hourly) data points in one day, or when they are far from model values (outside $3\varepsilon_t$). Similar observation quality controls have been applied in top-down studies (e.g., Chevallier et al., 2007). We used a series of sensitivity experiments (SR3 to SR6) to investigate the effect of adopting different uncertainty estimates and observation filters (Table S6). When we relax the observational filter to $5\varepsilon_t$, we infer a stronger biospheric sink: -0.85±0.50 PgC/yr (SR3) and -1.26±0.34 PgC/yr (SR4). Further relaxing the observational filter to, say, $8\varepsilon_t$ results in only small additional changes (<0.05 PgC/yr). When we assume a fixed observation error of 2 ppm, we infer a weaker biospheric sink: -0.68±0.52 PgC/yr (SR5) and -1.07±0.37 PgC/yr (SR6), almost the same with SR-1 and SR-2, respectively. These sensitivity tests demonstrate the qualitative robustness of the main result of the paper.

We also test the sensitivity of our *a posteriori* CO_2 fluxes to assumed *a priori* uncertainties. The inversions SR-7 and SR-8 correspond to inversions SR-1 and SR-2 but using an illustrative 20% reduction of the *a priori* uncertainty. *A posteriori* land biosphere fluxes for SR-7 (-0.55±0.45 PgC/yr) and SR-8 (-0.89±0.34 PgC/yr) are 17% and 20% weaker than values for SR-1 and SR-2, respectively, but consistent within their uncertainties. The direction of this change is as expected if we have better knowledge of the *a priori* flux estimates. The results of these sensitivity tests are also consistent with the main result of the paper.

Experiment #	**Inversion description**
SR-1	Uses NOAA obspack_co2_1_GLOBALVIEWplus_v3.2_2017-11-02 (GVI) database.
SR-2	As SR-1 and data from CMA, JR-STATION and WDCGG.
SR-3	As SR-1, but we assimilate all observations within 5 times of ε_t
SR-4	As SR-2, but we assimilate all observations within 5 times of ε_t
SR-5	As SR-1, but observation error is assumed to be a fixed value of 2 ppm.
SR-6	As SR-2, but observation error is assumed to be a fixed value of 2 ppm.
SR-7	As SR-5 but assuming 80% of a priori uncertainty.
SR-8	As SR-6 but assuming 80% of a priori uncertainty.

SR-9	As SR-1 but using ODIAC emissions from China.
SR-10	As SR-2 but using ODIAC emissions from China.
SR-11	An ensemble mean of 30 SR-2 inversions each with a random selection 5% of data removed across all sites and years.

Table S6 The inversions we present in this study.

The method we use to infer the land biosphere CO_2 flux is used widely in the community (Peylin et al.,2013; Thompson et al., 2016). First, we estimate net CO_2 fluxes by fitting our model to atmospheric CO_2 mole fraction data and then subtract fossil fuel emission estimates. Consequently, our estimate for the land biosphere CO_2 is sensitive to our assumed fossil fuel emissions. A recent study has developed a bottom-up inventory that revises downward Chinese fossil fuel emissions from 1997 to 2015 (Shan et al., 2018). In SR-1 and SR-2, we retain the spatial distribution of ODIAC fossil fuel emissions but scale the total emissions by an illustrative 88%. Comparisons between SR-1 and SR-9 and between SR-2 and SR-10, only differ by the magnitude of *a priori* fossil fuel emissions. Our land biosphere CO_2 flux for SR-1 (-0.66±0.52 PgC/yr) is 26% weaker than SR-9 (-0.89±0.52 PgC/yr), and SR-2 (-1.11±0.38 PgC/yr) is 25% weaker than SR-10 (-1.47±0.38 PgC/yr).

The CMA weekly flask and hourly continuous data we have reported represent collectively 36,939 individual points. To determine the importance of individual data points we ran estimated the carbon fluxes 30 times each with a different 5% of the data removed (SR-11). For each member of this ensemble, 1,847 randomly selected measurements (identified using a normal distribution) from across the network and across all years were removed. We find that the resulting nationwide carbon uptake estimate weakens by approximately 0.07 PgC/yr (6%) when we remove 5% of the data. The ensemble mean and median carbon flux estimate for China is -1.038 PgC/yr, ranging from -1.043 PgC/yr to -1.033 PgC/yr. The *a posteriori* flux estimates for the sub-continental regions (Figure S12) are similarly robust and approximately 6% weaker.

These results underscore the importance of being able to estimate independently fossil fuel and land biosphere fluxes of CO_2. On smaller geographical scales our *a posteriori* land biosphere fluxes for SWC are -0.35±0.15 PgC/yr (SR-2), -0.38±0.15 PgC/yr (SR-9) and -0.41±0.15 PgC/yr (SR-11), and for NEC are -0.05±0.14 PgC/yr (SR-2), -0.09±0.14 PgC/yr (SR-9), and -0.05±0.14 PgC/yr (SR-11), respectively.

Considering the results of all of our experiments, our *a posteriori* estimates for the CO_2 terrestrial biosphere uptake over China inferred using only the NOAA ESRL Obspack GLOBALVIEWplus data range from -0.55±0.52 PgC/yr to -0.89±0.52 PgC/yr. *A posteriori* estimates for the CO_2 terrestrial biosphere uptake over China that also include the CMA data range from -0.89±0.38 PgC/yr to -1.47±0.38 PgC/yr.

Evaluation

We evaluate our *a posteriori* atmospheric CO_2 values with Comprehensive Observation Network for TRace gases by AIrLiner (CONTRAIL) data and Total Carbon Column Observing Network (TCCON) data. First, we find neither data from

TCCON nor the CONTRAIL data are very sensitive to our *a posteriori* Chinese fluxes. Consequently, there is very little difference between *a posteriori* values from SR1 and SR2 and the independent data. It is because of this result that we used the satellite data as a way to evaluate the *a posteriori* fluxes (section 5) we inferred from the CMA *in situ* data.

Figure S9 shows that atmospheric CO_2 inferred from our different inversions (SR-1, SR-2 and an inversion using only GOSAT ACOS data), sampled at the location of CONTRAIL data, are very similar. This is because CONTRAIL data are not particularly sensitive to fresh Chinese outflow. Our comparison with CONTRAIL data uses exclusively ascent and descent measurements (500-900 hPa). These CONTRAIL data that recently been linked to changes in nearby upwind cities (Umezawa et al, 2020). So, variations in CONTRAIL data will also reflect changes in localized urban sources as well as larger scales relevant to our experiments.

We find that the TCCON site in Tsukuba (TK), Japan is the only TCCON site with any sensitivity to Chinese CO_2 fluxes, which is only modest. Figure S10 shows the comparison of these TCCON data and our *a posteriori* CO_2 concentrations sampled at TK. We find that our results from SR2 are only slightly better than SR1. The shift in the residual from late 2013 corresponds to a time when the Tsukuba Bruker 125HR was configured to include an additional IR channel and Si detector, allowing investigators to take correct a laser sampling error associated with the pre-processing of the GFIT spectral fitting method used in the GGG2014 data version (https://tccondata.org/2014). A laser sampling error correction that uses only the SWIR InGaAs detector spectra could help reduce the bias prior to January 2014.

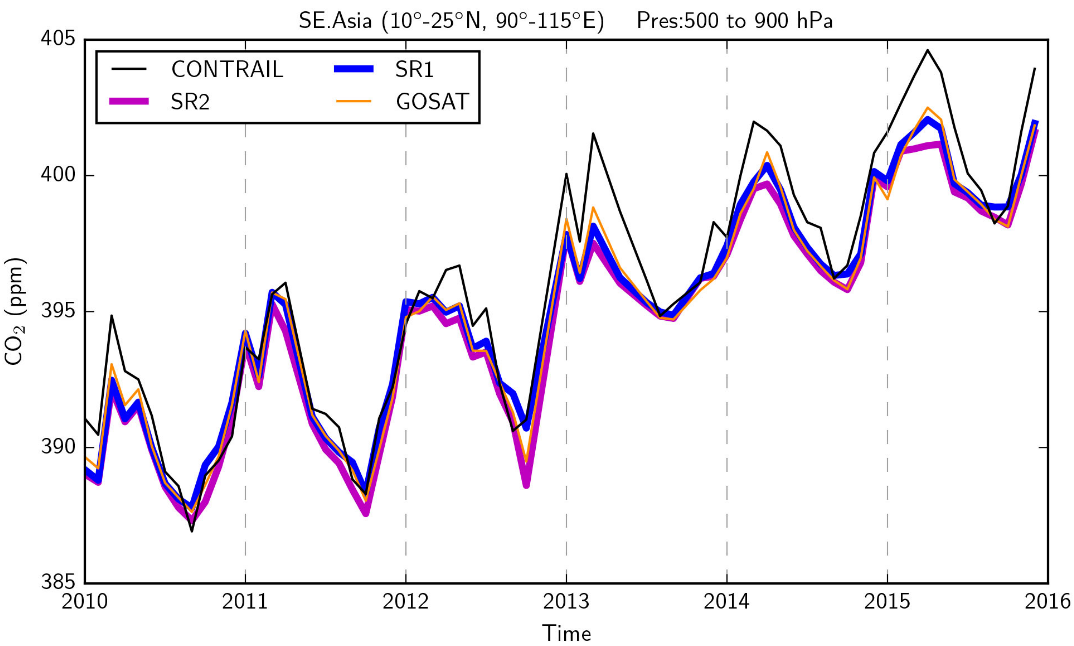

Figure S9 Comparison of *a posteriori* atmospheric CO_2 inferred from different inversions using subsets of atmospheric CO_2 mole fraction data collected over the region defined as 10°-25°N, 90°-115°E, 900-500 hPa by CONTRAIL data.

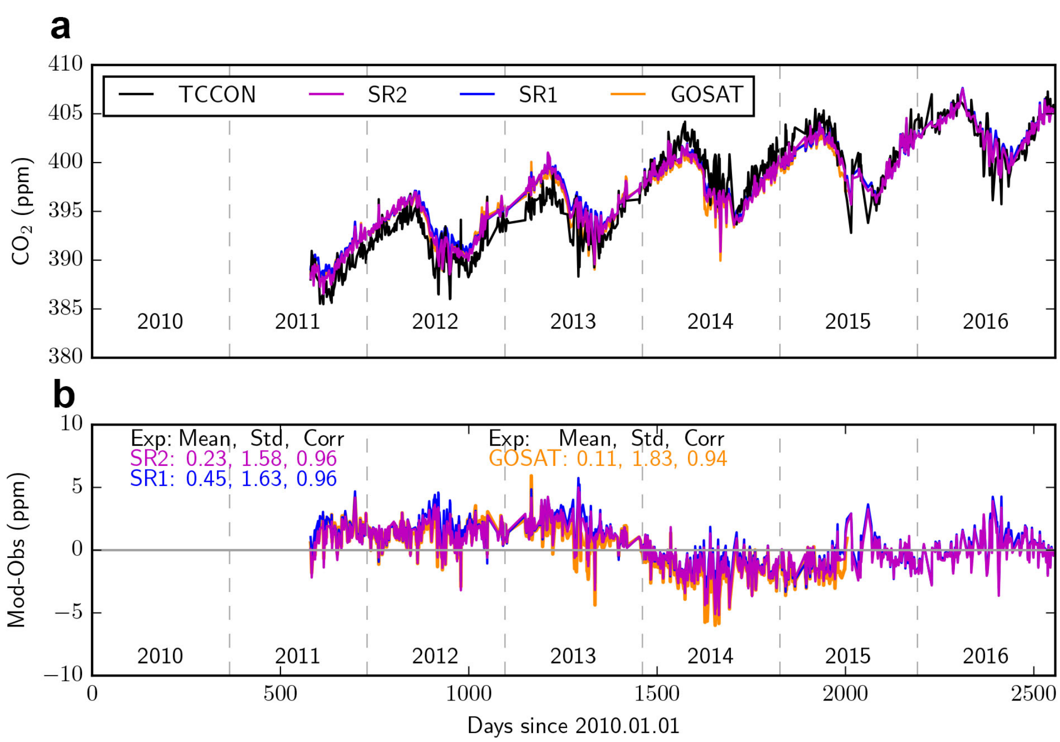

Figure S10 a Comparison of CO_2 column data observed by TCCON Tsukuba (TK) Japan and *a posteriori* atmospheric CO_2 columns inferred from different inversions that use subsets of atmospheric CO_2 mole fraction data, and **b** the corresponding model minus observation residuals.

Table S7 show select statistics of residuals between observed CO_2 mole fractions and model values corresponding to *a posteriori* flux estimates (SR-1 and SR-2). A *posteriori* CO_2 fluxes of SR-2 result in smaller mean residuals and smaller standard deviations about those means, relative to the observations compared to the SR-1 *a posteriori* CO_2 fluxes, as expected. The mean residuals of the 12 sites for SR-1 and SR-2 are -0.31 ppm and 0.04 ppm, respectively, and the corresponding standard deviations are 2.23 ppm and 2.03 ppm, respectively,

Site	**Country**	**Location (Lat,Lon)**	**Obs - posterior (SR-1) mean (ppm)**	**Obs-posterior (SR-1) 1-σ (ppm)**	**Obs-posterior (SR-2) mean (ppm)**	**Obs-posterior (SR-2) 1-σ (ppm)**
WLG	China	36.12°N,100.06°E	0	1.84	0.58	1.78
AKDL	China	47.10°N,87.93°E	1.09	2.33	1.14	1.79
JS	China,	29.63°N,114.22°E	-2.06	2.64	-1.08	2.33
SDZ	China	40.65°N,117.12°E	0.11	2.55	0.15	2.37
SL	China	28.00°N, 99.40°E	-1.59	2.56	0.56	2.30
LFS	China	44.73°N,127.60°E	0.50	3.54	0.33	2.68
LAN	China	30.03°N,119.72°E	-0.36	2.97	0.06	2.77
UUM	Mongolia	44.45°N,111.10°E	0.16	2.17	0.11	2.38

MNM	Japan	24.28°N,153.98°E	-0.14	0.90	-0.01	0.93
RYO	Japan	39.03N,141.82°E	-0.85	1.94	-1.04	1.86
TIK	Russia	71.60°N,128.89°E	0.15	0.64	0.12	0.53
DSI	Taiwan	20.70°N,116.73°E	-0.71	2.69	-0.42	2.68

Table S7 Mean statistics of the residuals between observed and *a posteriori* CO_2 mole fractions. 1-σ values denote the standard deviation about the mean values.

4 Forest Data over China

Here, we provide a critical review of correlative remote sensing and forest census data to put our result in a broader perspective. We focus on forest ecosystems because they represent the largest contribution to the net terrestrial carbon sink over China (Fang et al, 2007). We use satellite remote sensing data and national forest inventory data to examine forest change over China.

Remote sensing data of forest coverage and change

We use the MODIS Land Cover Type Yearly Climate Modeling Grid (CMG) product that has a spatial resolution of 0.05° (MCD12C1) and covers the time period 2001-2012 (Friedl et al, 2010). We use the 17 land cover classes defined by the International Geosphere Biosphere Programme (IGBP) global vegetation classification scheme. We estimate the forest change with MODIS landcover data (MCD12C1). All land cover types are classified as forest or non-forest.

Extended Data Figure 7 shows that most of China's forests are located in the south and northeast of the country. It also shows that forest cover (from non-forest to forest) has expanded considerably over the period 2001-2012, especially in SWC.

National forest inventories

China's State Forestry Administration (SFA) conducts a National Forest Inventory (NFI) every five years (Zeng et al., 2015). Each survey uses established international standard National forest resource continuous investigation techniques method. The SFA is currently in the process of the ninth NFI (2014-2018).

We use survey data from the first (1973-1976) to eighth (2009-2013) forest inventory data for the whole country and from the ninth forest survey for three southwest provinces: Yunnan, Guangxi, Guizhou. This is summarized in Extended Data Table 1. We focus on forest change on the country-scale, three provinces in SWC (Guangxi, Yunnan and Guizhou), and three provinces in NEC (Heilongjiang, Jilin and Liaoning).

Extended Data Figure 6 show how these surveys document the increase in forest coverage over China. Forest coverage in 2013 (22%) from the eighth survey has doubled since the 1970s (13%) from the first survey, increasing from 121.9 to 207.7 million hectares with corresponding forest stocks volume increasing from 8.6 to 15.1 billion cubic metres. This large-scale increase, particularly since the 1990s, is due to China's policy and investment on the protection of natural forest and afforestation and on the recovery of natural forest and large areas of plantation forest. In particular, since the start of the century forest resources have experience a period of rapid growth. All provinces show an increase in forest cover with Guizhou, Guangxi, and Liaoning changes the highest percentage increases.

We discuss data from the three most recent publicly available national surveys (6th, 7th, 8th) for the whole country, and also include results from the 9th survey for the three southwest provinces, Guangxi, Yunnan and Guizhou (Extended Data Table 1). From 1999-2013, Chinese forest area has increased by 22.2 million hectares and the plantation forest area increased by 16.1 million hectares. Forest stocks have increased by 2.7 billion m^3. This is due to Chinese forest protection and afforestation activities from 1999 to 2013.

The six provinces all have high forest coverage, especially Guangxi (60%) and Yunnan (59%). Yunnan and Heilongjiang have the largest forest stocks. For the three northeast provinces, Heilongjiang, Jilin and Liaoning, forest stocks have increased by 269.9, 106.1 and 75.7 million m^3, respectively. The increase of forest stock in Heilongjiang alone accounts for 10% of the whole country's growth.

The subtropical forest in SWC mainly comprises of evergreen broadleaf forest on the plains and coniferous forest in mountainous regions. Forest cover and forest area in SWC has progressively increased, especially the area associated with plantation forest. For the three provinces in Southwest China (Guangxi, Yunnan and Guizhou) over the period 2000 to 2016: forest coverage has increased by 18-20%, forest land area has increased by a factor of 1.4-1.8, and forest stock has increased by a factor of 1.4-2.2. The increase in forest stock Yunnan alone represents 19% of the national value.

A key factor for the rapid increase of forest in SWC is the increase of plantation forests. China currently has the world's largest afforestation area (FAO, 2016). SWC is one of the main regions of plantation (Huang et al., 2012). The Gunagxi Autonomous Region has the largest plantation area, representing approximately 9% of the total plantation within China [State Forestry Administration of the People's Republic of China's, 2018]. SWC includes the eucalyptus economic forests that have high growth rates. These eucalyptus forests represent 25% and 10% of planted forest in Guangxi and Yunnan. The plantation forest in Guangxi, Yunnan, and Guizhou accounts for a large proportion of the forest, the area proportion is 51%, 25% and 51% respectively and the stock proportion is 51%, 14% and 42% respectively in the 9th NFI.

The inventory data also show an increase of annual forest growth rate, from 3.6 m^3/year/ha (2003) to 4.2 m^3/year/ha (2013). Plantation forests grow more rapidly (5.5 m^3/year/ha) than natural forests (3.7 m^3/year/ha), the eucalyptus forest even can reach to 15 m^3/year/ha. Eucalyptus in Yunnan and Guangxi account for 50% of the whole country and is the second kind of plantation forest in Guangxi.

Another key factor that determines the magnitude of the carbon sink is the age of the forest. A consequence of the rapid increase of afforestation during the last 30 years is that Chinese forests contain a large fraction of young and mid-aged trees, which are associated with a high rate of carbon sequestration [Pan et al., 2004; Wang et al., 2011a; Yu et al, 2017]. Forests are classified as young, middle-aged, near-mature,

mature, and over-mature using distinct biological attributes, growth process, and management and utilization purpose [State Forestry Administration of the People's Republic of China's, 2018].

Extended Data Figure 9 shows that for China and the six provinces in SWC and NEC, typically 60% of forest areas are classified as young or middle-aged. In Jilin just over half of the forest area is young or middle-aged, and in Gunagxi and Guizhou over 80% of the forest areas is young or middle-aged.

Forests in Liaoning, Guizhou and Guangxi are the youngest, with 47%, 50%, and 46% classified young forest, respectively. From 1990 to 2010, forest coverage in Guizhou increased from 15% to 37%, Guangxi from 25% to 56%, and Liaoning from 27% to 38%. This reflects sustained afforestation, accompanied with high proportion of plantation forest in these three provinces. Extended Data Figure 9 shows these young forests represent only a small fraction of current forest stocks. This suggests that these young forests have still to realize their potential CO_2 sequestration capacity.

Remote sensing data of land biosphere activity

To interpret our *in situ a posteriori* CO_2 fluxes we use a variety of remote sensing data of land biosphere activity.

Leaf Area Index (LAI) is a vegetation index. It is defined as the one-sided green leaf area per unit ground area in broadleaf canopies and as half the total needle surface area per unit ground area in coniferous canopies. We use it here to track vegetation growth. We use MCD 15A2H:(MODIS/Terra+Aqua Leaf Area Index/FPAR 8-Day L4 Global 500 m SIN Grid V006) data (Myneni et al., 2015) to show the LAI information.

Normalized Difference Vegetation Index (NDVI) and Enhanced Vegetation Index (EVI) are alternative vegetative indices. They represent composite properties of leaf area, chlorophyll and canopy structure. We use MOD13C2 (MODIS/Terra Vegetation Indices Monthly L3 Global 0.05° CMG V006) (Didan et al.,2015) to get NDVI AND EVI information. The data are only retained with pixel reliability values masked as good data ("0") or marginal data ("1").

Gross Primary Productivity (GPP) is the amount of biomass that vegetation creates in a given length of time. We use it as a measure of the growth of terrestrial vegetation. The data product we use is determined on the radiation use efficiency concept. We use MOD17A2H (MODIS/TERRA Gross Primary Productivity 8-Day L4 Global 500 m SIN Grid V006) that is a cumulative 8-day composite of values with 500 metre pixel size based on the radiation-use efficiency concept (Running et al., 2015).

Net Photosynthesis (PSN) is the difference between GPP and the Maintenance Respiration (GPP-MR). The data we use are from the same files as GPP.

The MODIS data are downloaded from LP DAAC (Land Processes Distributed Active Archive Center, lpdaac.usgs.gov). The 500m resolution data are mosaicked and reprojected from the native sinusoidal projection to a geographic projection using the

MODIS Reprojection Tool (MRT). We used the MRT to re-project from the MODIS uniform grid (500m) to a geographical uniform grid (0.1°).

GOSAT Solar-Induced Fluorescence (SIF) is a measure of photosynthesis. Trees fluoresce in the range 650-800 nm, peaking at 685-690 nm and 730-740 nm. We use GOSAT SIF data retrieved using a spectral window centred on 755nm. Data are provided by University of Leicester on a regular 1°x1° grid (Somkuti et al, 2018).

Above-ground Biomass Carbon (ABC). We use ABC estimates (1993-2012) based on harmonized vegetative optical depth data for 1993 onwards derived from a series of passive microwave satellite sensors (Liu et al, 2015). These sensors include the Special Sensor Microwave Imager (SSM/I), Advanced Microwave Scanning Radiometer for Earth Observation System (AMSR-E), FengYun-3B Microwave Radiometer Imager (MWRI) and Windsat. Data are available from http://www.wenfo.org/wald/global-biomass/.

Extended Data Figure 8 shows multi-year means of these remote sensing data over China. NDVI, EVI, LAI and SIF data show values indicative of large biospheric activity over southern China (SWC and SEC) and NEC. These distributions of biospheric activity are consistent with the forest distribution showed in Extended Data Figure 7. As a result, the GPP and PSN are consistent with the distribution of NDVI, EVI, LAI and SIF. SWC has the largest PSN, which is consistent with our results shown in Figure 2 and Extended Data Figure 2. The ABC data show the above aground biomass carbon are mainly in SWC, CC and NEC, which illustrate the important role of these regions in carbon uptake in China, especially SWC and NEC.

Figure S11 show that NDVI and EVI over NEC and SWC have marginally increased from 2010-2016. NEC has a larger seasonal amplitude than SWC.

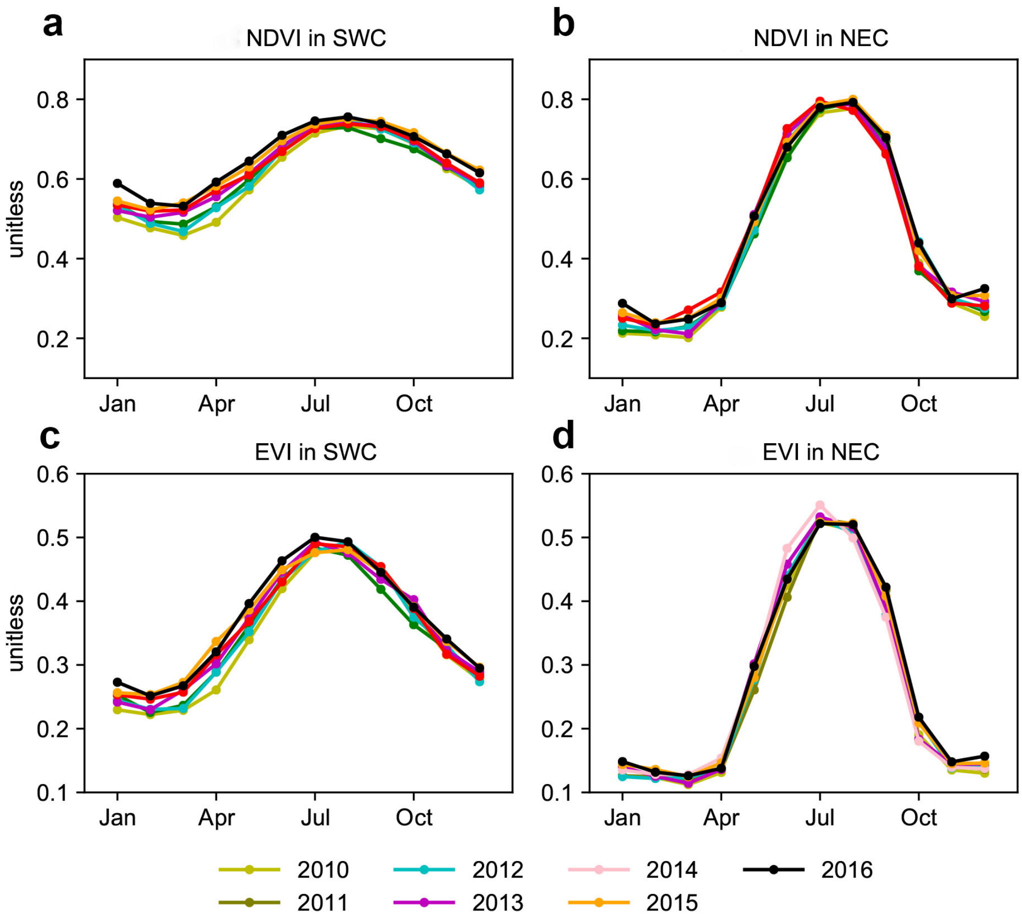

Figure S11 Seasonal variations in (top) NDVI and (bottom) EVI over (left) SWC and (right) NEC, 2010-2016.

Figure S12 shows year-to-year variations in NDVI from 2001 to 2016, including variations in the peak and trough values, mean annual values, and mean values during the growing season (April-November for SWC and SEC, and May-October for the other regions). We find a general increase of NDVI over China, especially the 'green' regions, including CC, EC, SWC, SEC and NEC. This is consistent with the increase in forest coverage over China, as discussed above. We also find that over TP, EC, SWC and SEC trough NDVI values have increased more than the peak values, resulting in a progressively dampened seasonal cycle. In contrast, over CC, NC, and NEC, we find that peak value has increased more than the trough value resulting in a larger seasonal cycle. We find similar results from our analysis of EVI (not shown).

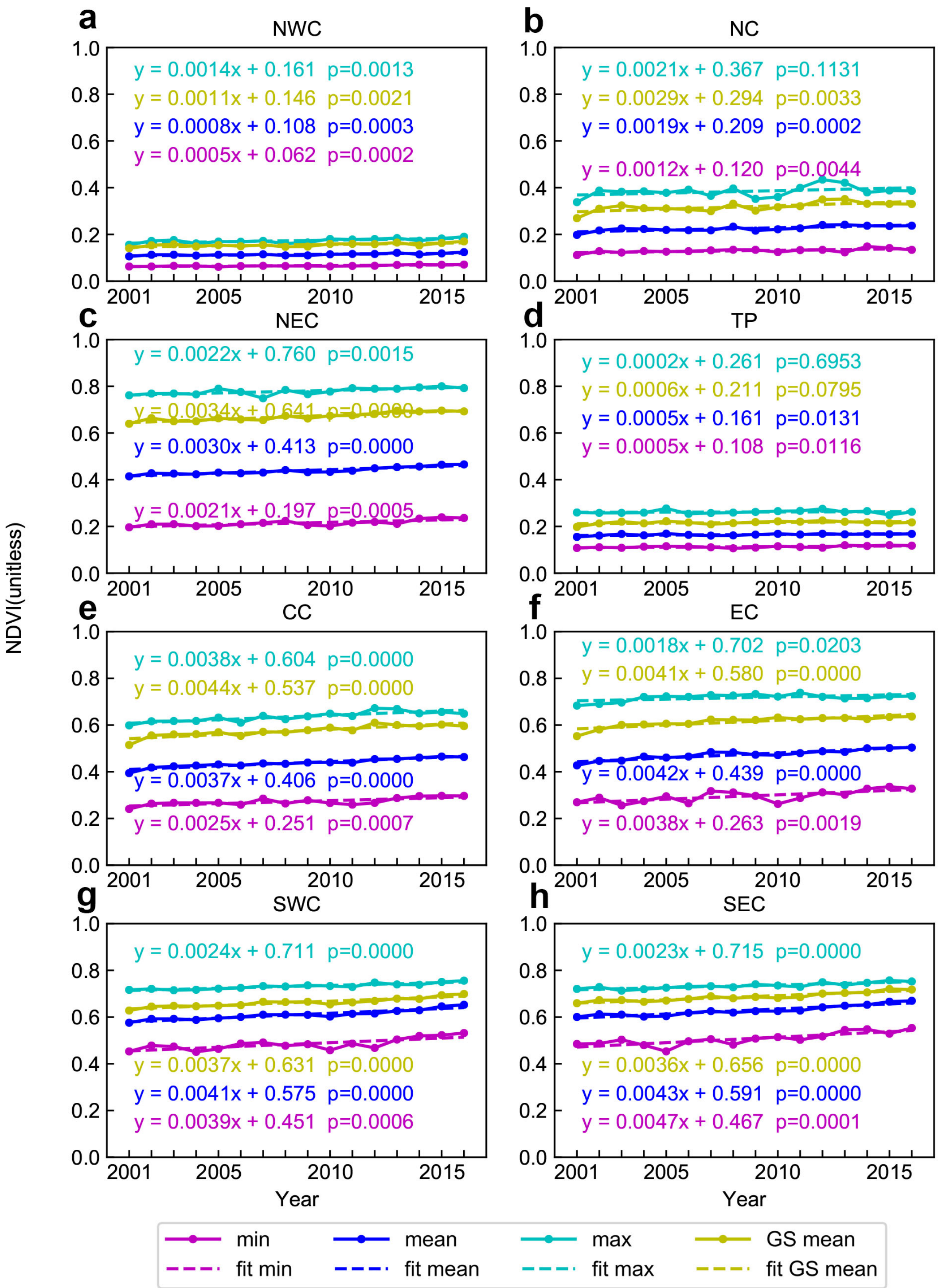

Figure S12 Year-to-year variations in NDVI over Chinese subcontinental regions (Extended Data Figure 2). Different colours denote the minimum and maximum values, mean annual value, and the mean value for the growing season. Inset is also shown the best-fit linear regression model and the associated statistical p-value.

Meteorology and hydrology measurements over SWC

We use GEOS-5 and GEOS-FP met fields to give the precipitation and temperature change in SWC (Figure S13).

Even though parts of SWC experienced two anomalous droughts during 2009-2015, autumn 2009 to spring of 2010 and during summer 2011 (Xu et al., 2015), we find an upward trend in precipitation over the broader geographical region.

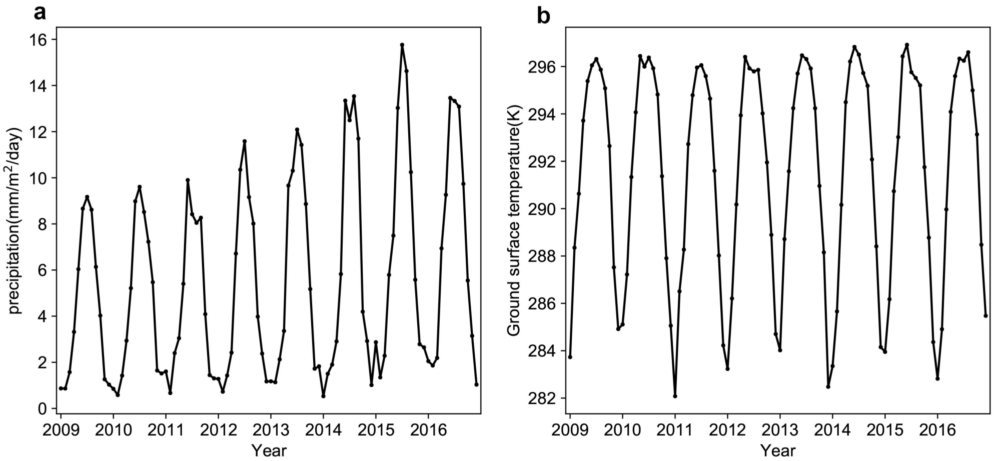

Figure S13 GEOS-5/GEOS-FP **a** precipitation rate (mm/m^2/day) and **b** surface skin temperature (K) over SWC from 2009 to 2016.

Gravity Recovery and climate experiment (GRACE) provides information about changes in the water column. Rooting depths of Chinese terrestrial ecosystems will likely be sufficiently deep that we cannot establish a direct and immediate relationship between vegetation and changes in precipitation. Changes in gravity due to changes in water column depth provide a much stronger relationship with vegetation access to water.

We use the surface mass change data based on the RL05 spherical harmonics from CSR (Center for Space Research at University of Texas, Austin), JPL (Jet Propulsion Laboratory) and GFZ (GeoforschungsZentrum Potsdam). The three different processing groups chose different parameters and solution strategies when deriving month-to-month gravity field variations from GRACE observations. We use the ensemble mean of the three data fields and multiply the data by the provided scaling grid. Data are available from http://grace.jpl.nasa.gov.

Analysis of GRACE data over SWC reveals a large seasonal cycle, determined by changes in precipitation, and a significant positive trend in the column water amount over this region. This trend also appears to be consistent with the increase in precipitation (Figure S14).

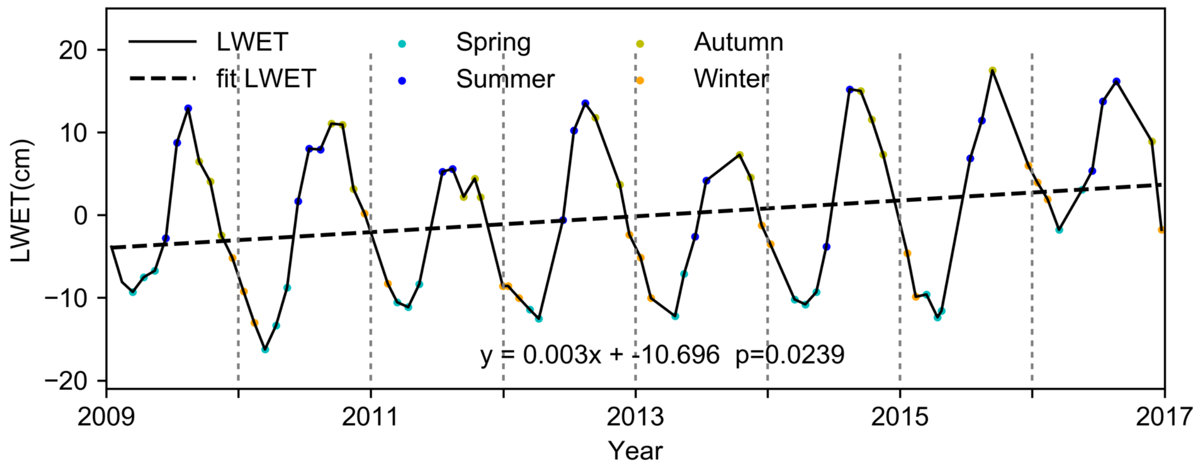

Figure S14 Anomalies in liquid water equivalent thickness (cm) inferred from the GRACE data over SWC. Anomalies are defined as the deviation from the 2004-2009 mean. The best fit regression line and the corresponding p-value are shown inset.

5 Comparison between *in situ* and satellite measurements inversion

As shown in Table 1 from the main paper, annual *a posteriori* biospheric CO_2 fluxes inferred from GOSAT and OCO-2 satellite column observations of CO_2 are less negative (smaller net sink) than annual fluxes inferred from the *in situ* (SR-2) data.

Extended Data Figure 5 shows monthly variations of the land biosphere flux inferred from *in situ* observations (inversion SR-2) and from GOSAT and OCO-2 satellite. We find that the higher annual fluxes inferred from GOSAT and OCO-2 are due mainly to higher *a posteriori* fluxes during October to April when data coverage is sparse (Extended Data Figure 4) and the fluxes are more influenced by *a priori* values.

Extended Data Figure 3 shows that mean *a posteriori* CO_2 flux distributions for May-September 2010-2015, inclusively, inferred from GOSAT and OCO-2 are consistent with the SR-2 inversion using *in situ* data. Uneven samplings during summer months (due to clouds and aerosols) result in some regional flux estimates reverting to a priori values (e.g. over the Tibetan Plateau), as expected.

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