

# Estimating Asian terrestrial carbon fluxes from CONTRAIL aircraft and surface CO<sub>2</sub> observations for the period 2006 to 2010

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## Abstract

Current estimates of the terrestrial carbon fluxes in Asia show large uncertainties particularly in the boreal and mid-latitudes and in China. In this paper, we present an updated carbon flux estimate for Asia ("Asia" refers to lands as far west as the Urals and is divided into Boreal Eurasia, Temperate Eurasia and tropical Asia based on TransCom regions) by introducing aircraft CO<sub>2</sub> measurements from the CONTRAIL (Comprehensive Observation Network for Trace gases by Airline) program into an inversion modeling system based on the CarbonTracker framework. We estimated the averaged annual total Asian terrestrial land CO<sub>2</sub> sink was about -1.56 Pg C yr<sup>-1</sup> over the period 2006-2010, which offsets about one-third of the fossil fuel emission from Asia (+4.15 Pg C yr<sup>-1</sup>). The uncertainty of the terrestrial uptake estimate was derived from a set of sensitivity tests and ranged from -1.07 to -1.80 Pg C yr<sup>-1</sup>, comparable to the formal Gaussian error of ±1.18 Pg C yr<sup>-1</sup> (1-sigma). The largest sink was found in forests, predominantly in coniferous forests (-0.64 Pg C yr<sup>-1</sup>) and mixed forests (-0.14 Pg C yr<sup>-1</sup>); and the second and third large carbon sinks were found in grass/shrub lands and crop lands, accounting for -0.44 Pg C yr<sup>-1</sup> and -0.20 Pg C yr<sup>-1</sup>, respectively. The peak-to-peak amplitude of inter-annual variability (IAV) was 0.57 Pg C yr<sup>-1</sup> ranging from -1.71 Pg C yr<sup>-1</sup> to -2.28 Pg C yr<sup>-1</sup>. The IAV analysis reveals that the Asian CO<sub>2</sub> sink was sensitive to climate variations, with the lowest uptake in 2010 concurrent with a summer flood and autumn drought and the largest CO<sub>2</sub> sink in 2009 owing to favorable temperature and plentiful precipitation conditions. We also found the inclusion of the CONTRAIL data in the inversion modeling system reduced the uncertainty by 11% over the whole Asian region, with a large reduction in the southeast of Boreal Eurasia, southeast of Temperate Eurasia and most Tropical Asian areas.

## 1. Introduction

The concentration of carbon dioxide (CO<sub>2</sub>) has been increasing steadily in the atmosphere since the industrial revolution, which is considered to be very likely responsible for the largest contribution of the climate warming (Huber and Knutti, 2011; Peters et al., 2011). Knowledge of the terrestrial carbon sources and sinks is critically important for understanding and projecting the future atmospheric CO<sub>2</sub> levels and climate change. The global terrestrial ecosystems absorbed about 1-3 Pg carbon every year during the 2000s, with obvious interannual variations, offsetting 10-40% of the anthropogenic emissions (Le Quéré et al., 2009; Maki et al., 2010; Saeki et al., 2013). However, estimates of the terrestrial carbon balance vary considerably when considering continental scales and smaller, as well as when estimating the CO<sub>2</sub> seasonal and inter-annual variability (Houghton, 2007; Peylin et al., 2013).

Asia, as one of the biggest northern hemisphere terrestrial carbon sinks, has a significant impact on the global carbon budget (Jiang et al., 2013; Patra et al., 2012; Piao et al., 2009; Piao et al., 2012; Peylin et al., 2013; Yu et al., 2013). It is estimated that Asian ecosystems contribute over 50% of the global net terrestrial ecosystem exchange (Maksyutov et al., 2003) and their future balance is thought to be a great source of uncertainty in the global carbon budget (Ichii et al., 2013; Oikawa and Ito, 2001). Even though the importance of the Asian ecosystems is increasingly recognized and many efforts have been carried out to estimate the Asian terrestrial carbon sources and sinks, they still remain poorly quantified (Ito, 2008; Patra et al., 2013; Patra et al., 2012; Piao et al., 2011). One reason is that a steep rise of fossil fuel emissions in most Asian countries has imposed large influences on the Asian CO<sub>2</sub> balance and leads to an increased variability of the regional carbon cycle (Francey et al., 2013; Le Quere et al., 2009; Patra et al., 2013; Patra et al., 2011; Raupach et al., 2007). In addition, rapid land-use change and climate change have likely increased the variability in the Asian terrestrial carbon balance (Cao et al., 2003; Patra et al., 2011; Yu et al., 2013). This makes it challenging to accurately estimate of CO<sub>2</sub> fluxes of the Asia ecosystems.

Currently two approaches are commonly used to estimate CO<sub>2</sub> fluxes at regional to global scales: the so-called “bottom-up” and “top-down” methods. The bottom-up

approach is based on local data or field measurements to retrieve the carbon fluxes, including direct measurements (Chen et al., 2012; Clark et al., 2001; Fang et al., 2001; Mizoguchi et al., 2009; Takahashi et al., 1999) and ecosystem modeling (Chen et al., 2007; Fan et al., 2012; Randall et al., 1996; Randerson et al., 1997; Sellers et al., 1986; Sellers et al., 1996). The top-down method uses atmospheric mole fraction data to derive the CO<sub>2</sub> sink/source information. As one of the important “top-down” approaches, atmospheric inverse modeling has been well developed and widely applied (Baker et al., 2006; Chevallier and O'Dell, 2013; Deng et al., 2007; Gurney et al., 2003; Gurney et al., 2004), and has shown to be particularly successful in estimating regional carbon flux for regions rich in atmospheric CO<sub>2</sub> observations like North America and Europe (Broquet et al., 2013; Deng et al., 2007; Peters et al., 2007; Peters et al., 2010; Peylin et al., 2013; Peylin et al., 2005; Rivier et al., 2011, 2010). However, estimating Asian CO<sub>2</sub> surface fluxes with inverse modeling remains challenging, and the inverted Asian CO<sub>2</sub> fluxes still exhibit a large uncertainty partly because of a lack of surface CO<sub>2</sub> observations. For example, in the TransCom3 annual mean control inversion, Gurney, *et al.* (2003) used a set of 17 models to estimate the carbon fluxes and obtained different results for the Asian biospheric CO<sub>2</sub> budget, ranging from a large CO<sub>2</sub> source of  $+1.00 \pm 0.61$  Pg C yr<sup>-1</sup> to a large sink of  $-1.50 \pm 0.67$  Pg C yr<sup>-1</sup> for the year 1992-1996. In the RECCAP (REgional Carbon Cycle Assessment and Processes) project, Piao, *et al.* (2012) presented the carbon balance of terrestrial ecosystems in East Asia from eight inversions during 1990-2009. The results from these eight inversion' models also show disagreement: six models estimate a net CO<sub>2</sub> uptake with the highest net carbon sink of  $-0.997$  Pg C yr<sup>-1</sup>, while two models show a net CO<sub>2</sub> source with the largest net carbon emission of  $+0.416$  Pg C yr<sup>-1</sup> in East Asia. The important role of the sparse observational network was demonstrated by Maki, *et al.* (2010), who reported a large Asian land sink of  $-1.17 \pm 0.50$  Pg C yr<sup>-1</sup> or much smaller sink of  $-0.65 \pm 0.49$  Pg C yr<sup>-1</sup> over the Asian region depending on which set of observations was included in the same inversion system. This situation suggests that a more accurate estimate of the surface CO<sub>2</sub> flux is urgently required in Asia, and the ability to base it on as much observational data as possible is key.

To expand the number of CO<sub>2</sub> observations, the aircraft project CONTRAIL has measured CO<sub>2</sub> mole fractions onboard passenger flights since 2005, and has produced

a large coverage of *in situ* CO<sub>2</sub> data ranging over various latitudes, longitudes, and altitudes (Machida et al., 2008; Matsueda et al., 2008). CONTRAIL observations have also already successfully been used to constrain surface flux estimates (Niwa et al., 2012; Niwa et al., 2011; Patra et al., 2011). Patra et al. (2011) reported the added value of CONTRAIL data to inform on tropical Asian carbon fluxes, as their signals are transported rapidly to the free troposphere over the west Pacific.

In this study, we also used the CONTRAIL CO<sub>2</sub> observations (<http://www.cger.nies.go.jp/contrail/>) together with a global network of surface observations to estimate the Asian weekly net ecosystem exchange of CO<sub>2</sub> (NEE) during the period 2006-2010. Our inversion model is the state-of-the-art CO<sub>2</sub> data assimilation system CTDAS (CarbonTracker Data Assimilation Shell, <http://carbontracker.eu/ctdas/>). Our work complements previous inverse modeling studies as it: (1) presents the inverted CO<sub>2</sub> results of Asian weekly net ecosystem exchange not shown previously; (2) uses surface observations not available in earlier top-down estimates; (3) assimilates the continuous CO<sub>2</sub> observation from a number of Asian continental sites for the first time; (4) includes extra free tropospheric CO<sub>2</sub> observations to further constrain the estimate; (5) uses a two-way atmospheric transport model TM5 (Krol et al., 2005) with higher horizontal resolution than previous global CO<sub>2</sub> data assimilation studies that focused on Asia (this study uses a 1×1 degree grid over Asia while globally a 2×3 degree resolution, see Figure 1b).

This paper is organized as follows. Methods and materials are described in Section 2, the inferred Asian land flux and its temporal-spatial variations are presented in Section 3. To examine the impact of CONTRAIL data on Asian flux estimates, we also compared inverse results with and without CONTRAIL data during 2006-2010. In Section 4, we compare our inverted Asian surface fluxes with previous findings and discuss our uncertainty estimates and future directions. Note that “Asia” refers to lands as far west as the Urals, and it is further divided into Boreal Eurasia, Temperate Eurasia and tropical Asia based on TransCom regions (Gurney et al., 2002; Gurney et al., 2003) (see small inset in the bottom left corner of Figure 1).

## 2. Methods and datasets

### 2.1 The atmospheric inversion model (CTDAS)

The atmospheric inverse model CTDAS was developed by NOAA-ESRL (National

Oceanic and Atmospheric Administration’s Earth System Research Laboratory) and Wageningen University, the Netherlands. Previous versions of the system have been applied successfully in North America and Europe (Masarie et al., 2011; Peters et al., 2007; Peters et al., 2010). CTDAS was designed to estimate net CO<sub>2</sub> terrestrial and oceanic surface fluxes by integrating atmospheric CO<sub>2</sub> concentration measurements, a global transport model, and a Bayesian synthesis technique that minimizes the difference between the simulated and observed CO<sub>2</sub> concentrations. The first step is the forecast of the atmospheric CO<sub>2</sub> concentrations using the transport model TM5 (Krol et al., 2005) with a global resolution of 3×2° and 1×1° over Asia (Figure 1b). The TM5 transport model is driven by meteorological data of the ERA-interim analysis of the European Centre for Medium-Range Weather Forecasts (ECMWF), and propagates four separate sets of bottom-up fluxes (details are presented in Section 2.2). The forecasted four-dimensional (4-D) concentrations ( $x, y, z, t$ ) are sampled at the location and time of the observed atmospheric CO<sub>2</sub> mole fractions, and subsequently compared. The difference between the observed and simulated CO<sub>2</sub> concentrations is minimized. This minimization of the mole fraction differences in CTDAS is done by tuning a set of linear scaling factors which are applied to find the set of sources and sinks that most closely match the observed CO<sub>2</sub> concentration in the atmosphere. As described in Peters *et al.* (2007), four a-priori and imposed CO<sub>2</sub> fluxes integrate in CTDAS to instantaneous CO<sub>2</sub> fluxes  $F(x, y, t)$  as follows:

$$F(x, y, t) = \lambda_r F_{bio}(x, y, t) + \lambda_r F_{oce}(x, y, t) + F_{ff}(x, y, t) + F_{fire}(x, y, t) \quad (1)$$

where  $F_{bio}$  and  $F_{oce}$  are 3-hourly, 1×1° a-priori terrestrial biosphere and ocean fluxes, respectively;  $F_{ff}$  and  $F_{fire}$  are monthly 1×1° prescribed fossil fuel and fire emissions, and  $\lambda_r$  is a set of weekly scaling factors, and each scaling factor is associated with a particular region of the global domain that is divided into 11 land and 30 ocean regions according to climate zone and continent. Nineteen ecosystem types (Olson et al., 1985) (Figure 1a) have been considered in each of 11 global land areas (Gurney et al., 2002), dividing the globe into 239 regions (239 = 11 land × 19 ecosystem types + 30 ocean regions). The actual region number assimilated in this system is 156, after excluding 83 regions which are associated with a non-existing ecosystem (such as “snowy conifers” in Africa). The corresponding scaling factors have been estimated

as the final product of CTDAS, and have been applied to obtain the terrestrial biosphere and ocean fluxes at the ecosystem and ocean basin scale by multiplying them with the a-priori fluxes. The adjusted fluxes are then put into the transport model to produce an optimized 4-D CO<sub>2</sub> mole fraction distribution.

## 5 **2.2 A-priori CO<sub>2</sub> flux data set**

In CTDAS, four types of CO<sub>2</sub> surface fluxes are considered: (1) the a-priori estimates of the oceanic CO<sub>2</sub> exchange are based on the air-sea CO<sub>2</sub> partial pressure differences from ocean inversions results (Jacobson et al., 2007). These air-sea partial pressure differences are combined with a gas transfer velocity computed from wind speeds in  
10 the atmospheric transport model to compute fluxes of carbon dioxide across the sea surface every 3 hours; (2) the a-priori terrestrial biosphere CO<sub>2</sub> fluxes are from GFED2 (Global Fire Emissions Database version 2), which is derived from the Carnegie-Ames Stanford Approach (CASA) biogeochemical modeling system (Van der Werf et al., 2006). A monthly varying NEE flux ( $NEE = R_e - GPP$ ) was  
15 constructed from two flux components: gross primary production ( $GPP$ ) and ecosystem respiration ( $R_e$ ), and interpolated to 3-hourly net land surface fluxes using a simple temperature  $Q_{10}$  relationship assuming a global  $Q_{10}$  value of 1.5 for respiration, and a linear scaling of photosynthesis with solar radiation. (3) The imposed fossil fuel emission estimates from the global total fossil fuel emission of the CDIAC (Carbon  
20 Dioxide Information and Analysis Center) (Marland et al., 2003) were spatially and temporally interpolated following the EDGAR (Emission Database for Global Atmospheric Research) database (Boden et al., 2011; Commission, 2009; Olivier and Berdowski, 2001; Thoning et al., 1989); (4) the biomass burning emissions are from GFED2, which combines monthly burned area information observed from satellites  
25 (Giglio et al., 2006) with the CASA biogeochemical model. Fire emissions in GFED2 are available only up to 2008, so for 2009 and 2010 we use a climatologically of monthly averages of the previous decade. Note that GFED3 (and now even GFED4) is available for quite a few years, and offers higher spatial resolutions in biomass burning emissions which are attractive for model simulation. But it uses a different  
30 product for the satellite observed NDVI and fPAR (MODIS instead of AVHRR) which causes a different seasonality in the biosphere fluxes which are calculated alongside the fire emissions in GFED, with a less realistic amplitude. Since this amplitude of the seasonal biosphere is important to us, we did not update to this new GFED3 product.



We also tested the GFED4 data with SIBCASA to make a new dataset of fire estimates but our analyses showed that the impact of using GFED4 versus GFED2 on estimated Asia fluxes is very weak.

### 2.3 Atmospheric CO<sub>2</sub> observations

5 In this study, two sets of atmospheric CO<sub>2</sub> observation data were assimilated: (1) surface CO<sub>2</sub> observations distributed by NOAA-ESRL (<http://www.esrl.noaa.gov/gmd/ccgg/obspack/>, data version 1.0.2) and by the WDCGG (World Data Centre for Greenhouse Gases, <http://ds.data.jma.go.jp/gmd/wdcgg/>) for the period 2006-2010 (the Asian surface site information is summarized in Figure 1a and the global surface sites *are in Supporting Information Appendix A*). Individual time series in this surface set were provided by many individual PIs which we kindly acknowledge; (2) for the free tropospheric CO<sub>2</sub> observations, we use the aircraft measurements from the CONTRAIL project for the period 2006-2010 (see Figure 1b).

15 A summary of Asian surface sites used in this study is shown in Table 1 and Figure 1a for reference. There are fourteen surface sites with over 7,957 observations located in Asia, including 10 surface flask observations and 4 surface continuous sites. The surface CO<sub>2</sub> mole fraction data used in this study are all calibrated against the same CO<sub>2</sub> standard (WMO-X2007). For most of the continuous sampling sites at the surface, we derived a daily averaged afternoon CO<sub>2</sub> concentration (12:00-16:00, Local time) for each day from the time series, while at mountain-top sites we construct an average based on nighttime hours (0:00-4:00, Local time) to reduce local influence and compare modeled with observed values only for well mixed conditions.

We note that from the CONTRAIL program (Machida et al., 2008; Matsueda et al., 25 2008), stratospheric CO<sub>2</sub> data were not included into CTDAS because the stratospheric observations had a seasonal phase shifting and its smaller amplitude was difficult to compare to the tropospheric measurements (Sawa et al., 2008). A summary of the CONTRAIL aircraft measurements is presented in Table 2 and Figure 1b. The CONTRAIL aircraft data are reported on the NIES 09 CO<sub>2</sub> scale, which are lower 30 than the WMO-X2007 CO<sub>2</sub> scale by 0.07 ppm at around 360 ppm and consistent in the range between 380 and 400 ppm (Machida et al., 2011). Thus the CONTRAIL CO<sub>2</sub> data sets are comparable to surface data. We follow the method from Niwa, *et al.*

(2012) to divide the data into 4 vertical bins (575–625, 465–525, 375–425, 225–275 hPa) from ascending and descending profiles and one vertical bin (225–275 hPa) from level cruising. We also divide CONTRAIL data into 42 horizontal bins/regions (Figure 1b), which amounts to a total of 65 bins. Before daily averaging the CONTRAIL measurements for each 65 regional/vertical bins, we pre-process the aircraft data to obtain free troposphere CO<sub>2</sub> values by filtering out the stratospheric CO<sub>2</sub> data using a threshold of potential vorticity (PV) > 2 PVU (1 PVU = 10<sup>-6</sup> m<sup>2</sup> s<sup>-1</sup> K kg<sup>-1</sup>), in which PV is calculated from TM5 (using ECMWF temperature, pressure and wind fields) (Sawa et al., 2008). A total number of 10,467 CO<sub>2</sub> aircraft observations over Asia have been used during January 2006 to December 2010 in our inversion.

#### 2.4 Sensitivity experiments and Uncertainty Estimation

Because the Gaussian uncertainties strongly depend on choices of prior errors in CTDAS, the formal covariance estimates for each week of optimization only reflect the random component of the inversion problem rather than a characterization of the true uncertainties of the estimated CO<sub>2</sub> flux. As an alternative, we performed a set of sensitivity experiments to obtain a more representative spread in the flux estimates and complement the formal Gaussian uncertainty estimates. We take different plausible alternative settings in CTDAS to design a more comprehensive sensitivity tests, and use the minimum and maximum flux inferred in these experiments to present the range of the true flux. Six inversions were performed to investigate the uncertainty span in this study:

Case 1: prior flux as in section 2.2 + observations as in section 2.3 + TM5 transport model runs at global 3×2° and a 1×1° nested grid over Asia. This is the base simulation (quoted as Surface-CONTRAIL) which is used to analyze the 5-year carbon balance in this study.

Case 2: same as Case 1, but excluding CONTRAIL observations. We use these results (quoted as Surface-Only) to examine the impact of CONTRAIL data on Asian flux estimates by comparison with Case 1.

Case 3: Like Case 1, but CTDAS runs with the updated fossil fuel emissions based on Wang, *et al.* (2012) over China. Different from fossil fuel data in Case 1, the data of Wang, *et al.* (2012) calculated carbon emissions from energy consumption, transportation, household energy consumption, commercial energy consumption,

industrial processes and waste. And the seasonal variations between the two datasets are different: the fossil fuel emissions in Case 1 had the largest carbon emission in January and the smallest carbon source in July every year, while data of Wang, et al. (2012) had the smallest fossil-fuel CO<sub>2</sub> emissions in February or March. This simulation is meant to partly address the impact of uncertainty in fossil fuel emissions over the region as suggested by Francey et al.,(2013)

Case 4: Like Case 1, but CTDAS runs based on 110% of prior biosphere flux derived from CASA-GFED2;

Case 5: Like Case 2, but the TM5 transport model is used at global 6°×4° without nested grids. This tests the impact of model resolution;

Case 6: Like case 2, but replacing the underlying land use map with MODIS data (Friedl et al., 2002) and keeping the number of ecoregions unchanged. The MODIS land use maps can be found in SI Appendix C.

The Cases 1 and 2 span the period 2006-2010 (2004-2005 were discarded as spin-up), while the other sensitivity experiments were done from 2008 to 2010 only when the observational coverage was best. In general, these six sensitivity tests investigate most variations in the components of the assimilation framework: prior fluxes, observations available, the ecoregion map, the fossil fuel emissions, and transport, and give alternative choices for the main components of the system. The sensitivity results are summarized in Table 3 and further discussed in the next section.

### 3. Results

We will from here on refer to carbon sinks with a negative sign, sources are positive, and will include the sign also when discussing anomalies (positive = less uptake or larger source, negative = more uptake or smaller source). We describe the results mainly over Asia (global flux estimates can be found in SI Appendix B), where we expected the CONTRAIL data to provide the additional constraints. Note that the results of Case 1 are analyzed as the best assimilation for the period of 2006-2010 in this study.

#### 3.1 CO<sub>2</sub> concentration simulations

First we checked the accuracy of the model simulation using the surface CO<sub>2</sub> concentration observations and CONTRAIL aircraft CO<sub>2</sub> measurements. Figure 2a

shows the comparison of modeled (both prior and posterior) CO<sub>2</sub> concentration with measurements at the discrete surface site of Mt. Waliguan (WLG, located at 36.29° N, 100.90° E). Note that the prior CO<sub>2</sub> concentrations here are not really based on a-priori fluxes only, as they are a forecast started from the CO<sub>2</sub> mixing ratio field that contains all the already optimized fluxes (1,..., n-1) that occurred before the current cycle of the data assimilation system (n). So these prior mole fractions only contain five weeks of recent un-optimized fluxes and constitute our ‘first-guess’ of atmospheric CO<sub>2</sub> for each site. For the WLG site, the comparison of the surface CO<sub>2</sub> time series shows that the modeled (both prior and posterior) CO<sub>2</sub> concentration is in general agreement with observed data during the period 2006-2010 (correlation coefficient R=0.87), although the modeled result still could not adequately reproduce all the observed CO<sub>2</sub> seasonal variations. The posterior annual model-observation mismatch of this distribution is  $-0.10 \pm 1.25$  ppm, with  $0.07 \pm 1.50$  ppm bias for the summer period (June-July-August) and  $0.02 \pm 0.80$  ppm bias for the winter period (December-January-February). The model-observation mismatch is a little larger in Case 2 without CONTRAIL data (model-observation mismatch:  $-0.13 \pm 1.26$  ppm), suggesting that the surface fluxes derived with CONTRAIL agree with the surface CO<sub>2</sub> mixing ratios at WLG station. Over the full study period, the WLG modeled mole fractions exhibit good agreement with the observed CO<sub>2</sub> time series and the changes in inferred mixing ratios/flux are within the specified uncertainties in our inversion system, an important prerequisite for a good flux estimate.

We also checked the inversion performance in the free troposphere in addition to the surface CO<sub>2</sub>. Figures 2b, 2c and 2d show the comparison between measured and modeled (both prior and posterior) mixing ratios in the free troposphere during the period 2006-2010 in the region covering 136-144°N, 32-40°E for 3 vertical bins (475-525, 375-425, 225-275 hPa). The observed vertical CO<sub>2</sub> patterns were reasonably reproduced by our model, with high correlation coefficient ( $R = 0.95, 0.94$  and  $0.93$  for 475-525, 375-425, 225-275 hPa, respectively) between CONTRAIL and (posterior) modeled CO<sub>2</sub>. The observed low vertical gradients for flight sections in 3 vertical bins (475-525, 375-425, 225-275 hPa) at northern mid-latitudes (32-40°E) were well captured by the model (both prior and posterior), indicating the transport model can reasonably produce the vertical structure of observations.

We found that the observed CO<sub>2</sub> concentration profiles were modeled better after

assimilation than before (modelled - observed =  $0.05 \pm 1.25$  ppm for a-priori and  $-0.01 \pm 1.18$  ppm for posterior), although our inverted (posterior) mole fractions still could not adequately reproduce the high values in winter (December-January-February) and the low values in summer (June-July-August). This mismatch of CO<sub>2</sub> seasonal amplitude suggests that our inverted (posterior) CO<sub>2</sub> surface fluxes do not catch the peak of terrestrial carbon exchange well. Previous studies have also found this seasonal mismatch, which may correlate with atmospheric transport, and has already been identified as a shortcoming in most inversions (Peylin et al., 2013; Saeki et al., 2013; Stephens et al., 2007; Yang et al., 2007). In addition, we found that the optimized CO<sub>2</sub> mole fractions seem better captured at low altitude with smaller standard deviations of the model-observation mismatch ( $\pm 1.12$ ,  $\pm 1.18$  and  $\pm 1.26$  ppm for 475-525, 375-425, 225-275 hPa) and higher correlation coefficient at 485-525 hPa. This suggests that the near surface layers are comparatively well constrained in CTDAS. Overall, the agreement between the model and measurements is fairly good and consistent with previously known behavior in the CarbonTracker systems, derived mostly from North American and European continuous sites. Note that all model-observation mismatch of Asian surface sites and CONTRAIL data have been included in Tables 1 and 2 (see column of " Bias (modeled) ").

## 3.2 Inverted Asian terrestrial CO<sub>2</sub> flux

### 3.2.1 Five-year mean

During the period 2006-2010, we found a mean net terrestrial land carbon uptake (a posteriori) in Asia of  $-1.56$  Pg C yr<sup>-1</sup>, consisting of  $-2.02$  Pg C yr<sup>-1</sup> uptake by the terrestrial biosphere and  $+0.47$  Pg C yr<sup>-1</sup> release by biomass burning (fire) emissions (Table 5). This terrestrial uptake compensates 38% of the estimated  $+4.15$  Pg C yr<sup>-1</sup> CO<sub>2</sub> emissions from fossil fuel burning and cement manufacturing in Asia. An uncertainty analysis for the Asian terrestrial CO<sub>2</sub> uptake derived from a set of sensitivity experiments has been conducted and put the estimated sink in a range from  $-1.07$  to  $-1.80$  Pg C yr<sup>-1</sup> (Table 3), while the 1-sigma of the formal Gaussian uncertainty estimate is  $\pm 1.18$  Pg C yr<sup>-1</sup> (Table 5). The estimated Asian net terrestrial CO<sub>2</sub> sink is further partitioned into a  $-1.02$  Pg C yr<sup>-1</sup> carbon sink in Boreal Eurasia and a  $-0.68$  Pg C yr<sup>-1</sup> carbon sink in Temperate Eurasia, with a  $+0.15$  Pg C yr<sup>-1</sup> CO<sub>2</sub> source in Tropical Asia.

The annual mean spatial distribution of net terrestrial carbon uptake over Asia is shown in Figure 3. Note that the estimated fluxes include terrestrial fluxes and biomass burning sources but exclude fossil fuel emissions. Most Asian regions were natural carbon sinks over the studied period, with strongest carbon uptake in the middle and high latitudes of the Northern Hemispheric part of Asia, while the low-latitude region releases CO<sub>2</sub> to the atmosphere. This flux distribution pattern is quite consistent with previous findings that northern temperate and high latitude ecosystems were large sinks (Hayes et al., 2011) and tropical land regions were carbon sources (Gurney et al., 2003).

The aggregated terrestrial CO<sub>2</sub> fluxes for 19 different ecosystems (Figure 1a) averaged over the period 2006-2010 are shown in Table 4 and Figure 4 (see Case 1). The majority of the carbon sink was found in the regions dominated by forests, crops and grass/shrubs. The largest uptake is by the forests with a mean sink of -0.77 Pg C yr<sup>-1</sup>, 83% of which (-0.64 Pg C yr<sup>-1</sup>) was taken up by conifer forests and 18% of which (-0.14 Pg C yr<sup>-1</sup>) by mixed forest, whereas the tropical forests released CO<sub>2</sub> (+0.08 Pg C yr<sup>-1</sup>). The estimated flux by CTDAS in Asian cropland ecosystems was -0.20 Pg C yr<sup>-1</sup>, with the largest crop carbon sink located in Temperate Eurasia (-0.17 Pg C yr<sup>-1</sup>). The grass/shrub lands in Asia absorbed -0.44 Pg C yr<sup>-1</sup>, with most of these grass/shrub sinks located in Temperate Eurasia (-0.36 Pg C yr<sup>-1</sup>). Other land-cover types (e.g. wetland, semi tundra and so on) sequestered about -0.15 Pg C yr<sup>-1</sup> (10% of total) over Asian regions. This suggests that according to our model, many ecosystems contributed to Asian CO<sub>2</sub> sinks, highlighting the complexity of the northern hemispheric total sinks.

### 3.2.2 Seasonal variability

Figure 5 shows the *prior* and *posterior* seasonal cycles of CO<sub>2</sub> fluxes for the Asia region and its three sub-regions as well as their Gaussian uncertainties. The seasonal amplitude in Boreal Eurasia as shown in Figure 5b proves to be the major contributor to the seasonal signal in Asia (Figure 5a). The large uptake of Eurasia Boreal occurs in summer and the large differences between the prior and the posterior fluxes are also found in the summer growing season, indicating the surface observation network and CONTRAIL data largely affect the estimated fluxes. Our monthly variability is very close to changes in Eurasia Boreal presented by Gurney, et al. (2004). In Figure 5c,

the seasonal pattern for the Eurasia temperate region shows a comparable pattern to Eurasia Boreal, but with a smaller seasonal magnitude. And the adjustments of the *prior* flux in spring and summer are also smaller. The largest CO<sub>2</sub> uptake in Eurasia Temperate subregion, however, is shifted from July to August compared to Boreal Eurasia, suggesting that a phase shift in the growing season occurred here with the highest CO<sub>2</sub> sink occurring later in the year. This seasonal cycle is slightly different from that reported by Gurney, et al. (2004), but shows a nice agreement with the seasonal dynamics of Niwa, et al. (2012) in the Southern Temperate Asia region, and of Patra et al. (2011) in the Northwest Asia region. In Tropical Asia (Figure 5d), the seasonal variation is very different from other Asian subregions characterized by a weak CO<sub>2</sub> uptake peak in August-October and much smaller carbon release in May-July. Overall, the *posterior* uncertainty reduction for the period 2006-2010 was about 25% in Asia, with the largest uncertainty remaining in the summer, suggesting that our model may not fully capture the biosphere sink signal in the growing season.

### 3.2.3 Interannual variability (IAV)

Figure 6 shows the estimated annual cumulative net ecosystem exchange in Asia during 2006-2010 as well as its anomaly with weekly intervals. Here, the biomass burning and fossil fuel emissions are excluded, and only the sum of fluxes from respiration and photosynthesis are shown, because biomass burning emissions have large interannual variability, especially for Tropical Asia.

The coefficient of IAV ( $IAV = \text{standard deviation}/\text{mean}$ ) in Asian land carbon flux is 0.12, with a peak-to-peak amplitude of 0.57 Pg C yr<sup>-1</sup> (amplitude = smallest - largest CO<sub>2</sub> sink), ranging from the smallest carbon uptake of -1.71 Pg C yr<sup>-1</sup> in 2010 and the largest CO<sub>2</sub> sink of -2.28 Pg C yr<sup>-1</sup> in 2009. As has been noted in many other studies (Gurney et al., 2008; Gurney et al., 2004; Mohammat et al., 2012; Patra et al., 2011; Peters et al., 2007; Peters et al., 2010; Yu et al., 2013), the IAV of the carbon flux strongly correlates with climate factors, such as air temperature, precipitation and moisture.

The year 2010 stands out as a particularly low uptake year in Asia, with a reduction of terrestrial uptake of 0.31 Pg C yr<sup>-1</sup> compared to the five-year mean. This reduction mainly appeared in Temperate Eurasia and Tropical Asia, leading to +0.25 Pg C yr<sup>-1</sup> (35% sink reduction) and +0.04 Pg C yr<sup>-1</sup> flux anomalies (24% sink reduction) in their

corresponding regions. In 2010, Asia experienced a set of anomalous climate events. For example, Temperate Eurasia experienced a severe spring/autumn drought, a heavy summer flood and a heat wave occurred in 2010 (NationalClimateCenter, 2011). From Figure 6b, we can see that 2010 did not show large anomalies until after the spring growing season. As anomalous climate appeared, the summer flood and autumn drought were identified as dominant climatic factors controlling vegetation growth and exhibiting a significant correlation with the land carbon sink, particularly in the croplands, grasslands and forests of Temperate Eurasia. In the end, 2010 only showed  $-1.71 \text{ Pg C yr}^{-1}$  biospheric  $\text{CO}_2$  uptakes (excluding fires) by the end of the year.

In contrast to 2010, the year 2009 had the strongest carbon sink for the study period, with much stronger uptake in Temperate Eurasia ( $-0.20 \text{ Pg C yr}^{-1}$  anomaly, 28% increase in  $\text{CO}_2$  uptake) as well as in Boreal Eurasia ( $-0.05 \text{ Pg C yr}^{-1}$  anomaly, 4% uptake increase compared to the five-year mean). It can be seen that 2009 started with a lower-than-average release of carbon in the first 4 months (17 weeks) of the year amounting to  $+0.28 \text{ Pg C yr}^{-1}$  compared to the five-year average of  $+0.45 \text{ Pg C yr}^{-1}$ . This variation of the Asian terrestrial carbon sink in the spring vegetation growing season may partly relate to a higher spring temperature in 2009 which induced an earlier onset of the growing season and led to a high vegetation productivity by extending the growing season (Mohammat et al., 2012; Richardson et al., 2009; Walther et al., 2002; Wang et al., 2011; Yu et al., 2013). From Figure 6b, 2009 shows a very high carbon uptake in the summer growing season (June-August, week 22 to 32) concurrent with favorable temperature and abundant precipitation conditions. After this summer, the vegetation productivity returned back to normal and the total cumulative carbon sink added up to  $-2.28 \text{ Pg C yr}^{-1}$  at the end of the year with  $-0.26 \text{ Pg C yr}^{-1}$  extra uptake compared to the five-year mean.

### 3.2.4 Uncertainty Estimation

Table 3 presents the estimated annual mean NEE across the alternative sensitivity experiments. The time spans are different among these 6 tests: Case 1 (surface-CONTRAIL) and Case 2 (surface-Only) run for the period 2006-2010 (2000-2005 servers as a spin-up period), while Cases 3 to 6 run for the period 2008-2010. To compare other alternative sensitivity estimates for the same period from 2008 to 2010,



we calculated this three-year average of annual Asia CO<sub>2</sub> fluxes (2008-2010) from all the 6 tests to be -1.61, -1.15, -1.69, -1.80, -1.23 and -1.07 PgC yr<sup>-1</sup>, respectively. The Asian CO<sub>2</sub> uptake thus ranges from -1.07 to -1.80 Pg C yr<sup>-1</sup> across our sensitivity experiments, which complements the Gaussian error. Despite the small numbers of years included, this range suggests that the Asian terrestrial was a sizable sink, while a carbon source implied in previous studies by the 1-sigma Gaussian error of ±1.18 Pg C yr<sup>-1</sup> on the estimated mean, is very unlikely. The largest sensitivity in inferred flux is to the change of prior terrestrial biosphere fluxes (Case 4, difference = Case 4 - Case 1). The inversions with different model resolutions (Case 5, difference = Case 5 - Case 2) and with different Chinese fossil fuel emissions (Case 3, difference = Case 4 - Case 1) also show large variations in the inverted CO<sub>2</sub> fluxes, while the sensitivity to the change of land cover types (Case 6, difference = Case 6 - Case 2) is generally modest. This highlights the current uncertainties in the Asian sink and the best method to estimate it from inverse modeling.

### 3.2.5 Impacts of the CONTRAIL data on inverted Asian CO<sub>2</sub> Flux

We examined the impacts of the CONTRAIL data on Asian flux estimation by comparing results from Case 1 (Surface-CONTRAIL) and Case 2 (Surface-Only)(Table 5 and Figure 7a). Note that the uncertainties shown in the Table 5 and Figure 7b are now the Gaussian uncertainties as we did not repeat all sensitivity experiments. As shown in Table 5, inclusion of the CONTRAIL data induces an averaged extra CO<sub>2</sub> sink of about -0.47 Pg C yr<sup>-1</sup> to Case 1 (0.47 = 1.56 - 1.09), with most addition to the grass/shrub ecosystem (Figure 4). The spatial pattern of Asian fluxes also changed considerably (see Figure 7a). For instance, a decrease in CO<sub>2</sub> uptake was found in the northern area of Boreal Eurasia together with an increase in the south of Boreal Eurasia, leading to almost identical total carbon sink strength in Boreal Asia between with and without CONTRAIL data. Whereas the estimated flux distribution in Tropical Asia showed a small spatial change and a large increase in regional sink size with CONTRAIL observations included.

Table 5 and Figure 7b shows the reduction of the Gaussian error between Case 1 and Case 2. The error reduction rate (ER) is calculated as a percentage:

$$ER = (\sigma_{Surface-Only} - \sigma_{Surface-CONTRAIL}) / \sigma_{Surface-Only} \times 100, \quad (2)$$

Where  $\sigma_{Surface-Only}$  and  $\sigma_{Surface-CONTRAIL}$  are Gaussian errors in Case 2 (Surface-Only) and Case 1 (Surface-CONTRAIL), respectively. By including the additional CONTRAIL data into the inversion system, the uncertainty of the posterior flux over Asia is significantly reduced ( $>10\%$ ), especially for the southeast of Boreal Eurasia, southeast of Temperate Eurasia and Tropical areas (up to 20-30%). The more pronounced reduction was found in Boreal Eurasia and Tropical Asia (reducing by 14% and 15%, respectively). This suggests that current surface CO<sub>2</sub> observations data alone do not sufficiently constrain these regional flux estimation (there are no observation sites in Boreal Eurasia and only one in Tropical Asia), and the additional CONTRAIL CO<sub>2</sub> observations impose an extra constraint that can help reduce uncertainty on inferred Asia CO<sub>2</sub> fluxes, especially for these two surface observation sparse regions.

#### 4. Discussions and Conclusions

##### 4.1 Impact of CONTRAIL

Our modeling experiments reveal that the extra aircraft observations shift the inverted CO<sub>2</sub> flux estimates by imposing further constraints. This confirms the earlier findings by Saeki, *et al.* (2003) and Maksyutov, *et al.* (2013) that the inverted fluxes were sensitive to observation data used. For Tropical Asia, inclusion of the CONTRAIL data notably reduced the uncertainties (about 15% reduction). Compared with an inversion study with the CONTRAIL data for the Tropical Asia region (Niwa *et al.*, 2012), the error reduction rate in land flux estimation in this study for the same region is smaller than that of Niwa, *et al.* (34%). This difference in uncertainty reduction likely results from the differences in inversion system design between these two studies, of which vertical mixing represented in transport model, and covariance assigned to prior fluxes are typically most important. We furthermore note that the set of observations used in these studies was not identical, we for instance included one tropical surface site (BKT, see Table 1 and Figure 1a) to constrain the inferred flux estimation but Niwa, *et al.* (2012) did not.

Our results share other features with the Niwa *et al.* (2012) study, for instance the largest impact on the least data constrained regions. As reported by Niwa, *et al.* (2012), the inclusion of CONTRAIL measurements not only constrains the nearby fluxes, but also reduces inferred flux errors in the regions far from the CONTRAIL

measurement locations. For instance, in Boreal Eurasia, where no surface site exists and which is far from the CONTRAIL data locations (after pre-processing of horizontal/vertical bins and filter operation of stratospheric, there is no CONTRAIL observation available over this region), uncertainty reductions are large (14%  
5 reduction in uncertainty). Similar results were also presented by Niwa, *et al.* (2012), with an 18% error reduction in Boreal Eurasia. These two studies consistently suggest that including the CONTRAIL measurements in inversion modeling systems will help to increase the NEE estimation accuracy over Boreal Eurasia.

The CONTRAIL constraint on Temperate Eurasia is generally modest, only having a  
10 6% error reduction. This may be because Temperate Eurasia has more surface observation sites than other regions in Asia. However, it is interesting that the difference in inverted NEE in this region between Surface-Only and Surface-CONTRAIL is large ( $-0.35 \text{ Pg C yr}^{-1}$ ), but inconsistent with Niwa *et al.* (2012). One cause of this is likely the sensitivity of these inverse systems to vertical transport  
15 (Stephens *et al.*, 2007), as also suggested by Niwa, *et al.*(2012). The uneven distribution of observations at the surface and free troposphere may also aggravate this discrepancy.

#### **4.1 Comparison of the estimated Asian CO<sub>2</sub> Flux with other studies**

Our estimated Asian terrestrial carbon sink is about  $-1.56 \text{ Pg C yr}^{-1}$  for the period  
20 2006-2010. Most parts of Asian were estimated to be CO<sub>2</sub> sinks, with the largest carbon sink ( $-1.02 \text{ Pg C yr}^{-1}$ ) in Boreal Eurasia, a second large CO<sub>2</sub> sink ( $-0.68 \text{ Pg C yr}^{-1}$ ) in Temperate Eurasia, and a small source ( $+0.15 \text{ Pg C yr}^{-1}$ ) in Tropical Asia. This spatial distribution of estimated terrestrial CO<sub>2</sub> fluxes is overall comparable to the results for the period of 2000 to 2009 by Saeki *et al.* (2013), derived from an  
25 inversion approach focusing on Siberia with additional Siberian aircraft and tower CO<sub>2</sub> measurements, especially in the high latitude areas.

Comparisons of our inverted CO<sub>2</sub> flux with previous studies are summarized in Table 6. In Boreal Eurasia, our inferred land flux ( $-1.02 \text{ Pg C yr}^{-1}$ ) is higher than Gurney *et al.* (2003) ( $-0.59 \text{ Pg C yr}^{-1}$  during 1992-1996), but close to Maki *et al.* (2010) ( $-1.46$   
30  $\text{Pg C yr}^{-1}$  during 2001-2007), CTE2013 ( $-0.93 \text{ Pg C yr}^{-1}$ ) and CT2011\_oi ( $-1.00 \text{ Pg C yr}^{-1}$ , downloaded from <http://carbontracker.noaa.gov>). In Temperate Eurasia, our inverted flux is  $-0.68 \text{ Pg C yr}^{-1}$ , which is well consistent with Gurney *et al.* (2003) ( $-0.60 \text{ Pg C yr}^{-1}$ ), but higher than CTE2013 ( $-0.33 \text{ Pg C yr}^{-1}$ ) and CT2011\_oi ( $-0.41$

Pg C yr<sup>-1</sup>) even though we used a similar inversion framework. One reason of this discrepancy is likely that different zoomed regions were configured in the inversion system. Another main factor is likely the inclusion of CONTRAIL largely impacts on our Temperate Eurasia's carbon estimates. In Tropical Asia, our estimate is +0.15 Pg C yr<sup>-1</sup>, which is in the range of Niwa *et al.*(2012) (+0.45 Pg C yr<sup>-1</sup>, GVCT) and Patra *et al.*(2013) (-0.104 Pg C yr<sup>-1</sup>), both including aircraft CO<sub>2</sub> measurements in their inversion modeling, and very close to the CTE2013 (+0.22 Pg C yr<sup>-1</sup>) and CT2011\_oi (+0.14 Pg C yr<sup>-1</sup>). The estimated total Asian terrestrial carbon sink is -1.56 Pg C yr<sup>-1</sup>, which is close to the CTE2013 (-1.05 Pg C yr<sup>-1</sup>) and CT2011\_oi (-1.27 Pg C yr<sup>-1</sup>).

10 The IAVs comparison between the results from this study and from CTE2013 is also presented in Table 7 (different from IAV in Section 3.2.2, these results include biomass burning emissions). The IAVs are different between inferred terrestrial CO<sub>2</sub> flux of this study and CTE2013. In Boreal Eurasia, there was a moderate Asian CO<sub>2</sub> sink in 2007 for CTE2013, while the results from this study show the highest carbon uptake for this year; in CTE2013, the strongest terrestrial CO<sub>2</sub> sink occurs in 2008, while from our estimates the sink in 2008 was weaker than that in 2007. For Eurasia temperate, the highest land sink occurs in 2007 for CTE2013, while in this study, the highest occurs in 2009. In Tropical Asia, there is very similar IAVs between CTE2013 and this study, but the size of carbon sink is inconsistent. Differences likely stems from the additions of Asian sites and CONTRAIL data in this study. Compared to previous findings, our updated estimation with these additional data seems to support a larger Asian carbon sink over the past decade.

The spatial patterns of NEE in Asia are complex because of large land surface heterogeneity, such as land cover, vegetation growth rates, and soil types, and varying responses to climate variations. This makes accurately estimating NEE over Asia challenging. We believe this study is therefore useful to improve our understanding of the Asia regional terrestrial carbon cycle even though our estimation still has remaining uncertainties and biases in the inverted fluxes. By these comparisons, we can also conclude that our inferred Asia land surface CO<sub>2</sub> fluxes support a view that both large boreal and mid-latitude carbon sinks in Asian are balanced partly by a small tropical source. This would support the earlier suggestion that Asia is of key interest to better understand the global terrestrial carbon budget in the context of climate change.

The majority of the CO<sub>2</sub> sink was found in the areas dominated by forests, crops and grass/shrubs. Asian forests were estimated to be a large sink (-0.77 Pg C yr<sup>-1</sup>) during 2006-2010, the sink size is slightly larger than the bottom-up derived results of Pan, *et al.* (2011) ( -0.62 Pg C yr<sup>-1</sup>) for the period 1990-2007. One cause of this discrepancy is likely due to that our estimate is presented at a coarse resolution (a 1×1° grid may contain other biomes with lower carbon uptake than forests). Another reason may be that about half of Temperate Eurasia was not included in the statistical analysis by Pan *et al.* (2011). Note that the carbon accumulation in wood products is not considered in our estimates and needs further analysis in future studies.

10 The croplands in Asia were identified to be an average sink of -0.20 Pg C yr<sup>-1</sup> during 2006-2010. The uptake in croplands is likely associated with agricultural technique and crop management. Different from other natural ecosystems, crop ecosystems are usually under intensive farming cultivation, with regular fertilizing and irrigation of the crops. This increases crop production, and in return leads to high residues and root  
15 to the soil, which increases the carbon sink in cropland (Chen *et al.*, 2013). However, the accumulation of crop carbon in most crop ecosystems is relatively low, and agricultural areas are even considered not to contribution to a long-term net sink (Fang *et al.*, 2007; Piao *et al.*, 2009; Tian *et al.*, 2011). This is because the carbon accumulation in the crop biomass is harvested at least once per year and released back  
20 as CO<sub>2</sub> to the atmosphere after consumption. We should note that our estimate in the crop sink is different from the results of “crop no contribution ” (Piao *et al.*, 2009). Our atmospheric inversion system can well capture the crop’s strong CO<sub>2</sub> uptake during the growing season, but the atmosphere locally does not reflect the emission of the harvested crops, which normally has been transported laterally and is consumed  
25 elsewhere. This harvested product is likely released from a region with high population density and hard to detect against high fossil fuel emissions, whereas the estimated crop flux remains a large net CO<sub>2</sub> uptake over the period considered even though the crop flux into the soil is relatively small. Thus the croplands’ sink in this study might be overestimated due to the absence of harvesting in our modeling  
30 system. This issue was also raised by Peters *et al.* (2007; 2010).

Grassland/Shrub ecosystems also play an important role in the global carbon cycle, accounting for about 20% of total terrestrial production and could be a potential carbon sink in future (Scurlock and Hall, 1998). The grass/shrub lands in Asia

absorbed a total of  $-0.44 \text{ Pg C yr}^{-1}$ , accounting for about 25% of the total Asian terrestrial  $\text{CO}_2$  sink, which is close to the averaged global grassland sink percentage of 20%. Compared to the bottom-up results that net ecosystem productivity was  $10.18 \text{ g C m}^{-2} \text{ yr}^{-1}$  by Yu, *et al.* (2013), our estimate of  $34.32 \text{ g C m}^{-2} \text{ yr}^{-1}$  is much higher. This  
5 might due to that the areas in this study include shrubs whereas other studies only considering grass lands.

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**Tables and figure captions**

**Tables**

**Table 1** Summary of the 14 Asian surface CO<sub>2</sub> observation sites assimilated between January 1, 2006 and December 31, 2010. The frequency of continuous data is one per day (when available), while discrete surface data point is generally available once per week. MDM (model-data-mismatch) is a value assigned to a given site that is meant to quantify our expected ability to simulate observations and used to calculate the innovation X<sup>2</sup> (Inn. X<sup>2</sup>) statistics. N denotes the number of observations used in CTDAS. Flagged observations refer to a model-minus-observation difference that exceeds 3 times the model-data-mismatch, these observations are therefore excluded from assimilation. The bias is the average from posterior residuals (assimilated values - measured values), while the modeled bias is the average from prior residuals (modeled values - measured values)

| Site  | Name                       | Lat, Lon, Elev.        | Lab      | N(flagged) | MDM | Inn. X <sup>2</sup> | Bias(modeled) |
|-------|----------------------------|------------------------|----------|------------|-----|---------------------|---------------|
| 1 WLG | Waliguan,China             | 36.29°N,100.90°E,3810m | CMA/ESRL | 254(19)    | 1.5 | 0.83                | -0.10(-0.14)  |
| 2 BKT | Bukit Kototabang,Indonesia | 0.20°S,100.312°E,864m  | ESRL     | 172(0)     | 7.5 | 0.73                | 5.53(5.51)    |
| 3 WIS | Sede Boker,Israel          | 31.13°N,34.88°E,400m   | ESRL     | 239(1)     | 2.5 | 0.62                | -0.10(-0.15)  |
| 4 KZD | Sary Taukum,Kazakhstan     | 44.45°N,77.57°E,412m   | ESRL     | 167(6)     | 2.5 | 1.16                | -0.08(0.50)   |

|        |                         |                          |          |         |     |      |              |
|--------|-------------------------|--------------------------|----------|---------|-----|------|--------------|
| 5 KZM  | Plateau Assy,Kazakhstan | 43.25° N ,77.88° E,2519m | ESRL     | 155(2)  | 2.5 | 0.96 | 0.50(0.63)   |
| 6 TAP  | Tae-ahn Peninsula,Korea | 36.73° N,126.13° E,20m   | ESRL     | 181(3)  | 7.5 | 0.60 | 1.82(2.13)   |
| 7 UUM  | Ulaan Uul,Mongolia      | 44.45° N,111.10° E,914m  | ESRL     | 231(5)  | 2.5 | 1.17 | 0.10(0.28)   |
| 8 CRI  | Cape Rama,India         | 15.08° N,73.83° E,60m    | CSIRO    | 33(1)   | 3   | 1.40 | -1.97(-2.11) |
| 9 LLN  | Lulin,Taiwan            | 23.47° N,120.87° E,2862m | ESRL     | 220(20) | 7.5 | 0.99 | 2.62(2.65)   |
| 10 SDZ | Shangdianzi, China      | 40.39° N,117.07° E,287m  | CMA/ESRL | 60(15)  | 3   | 1.18 | 0.15(0.18)   |

**continuous samples in Asia:**

|        |                          |                         |      |           |   |      |              |
|--------|--------------------------|-------------------------|------|-----------|---|------|--------------|
| 11 MNM | Minamitorishima,Japan    | 24.29° N,153.98° E,8m   | JMA  | 1624(0)   | 3 | 0.76 | 0.15(0.16)   |
| 12 RYO | Ryori,Japan              | 39.03° N,141.82° E,260m | JMA  | 1663(48)  | 3 | 0.90 | 0.46(0.69)   |
| 13 YON | Yonagunijima, Japan      | 24.47° N,123.02° E,30m  | JMA  | 1684(3)   | 3 | 0.78 | 1.53(1.67)   |
| 14 GSN | Gosan, Republic of Korea | 33.15° N,126.12° E,72m  | NIER | 1274(109) | 3 | 1.99 | -1.01(-0.82) |

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**Table 2** Summary of the Asian CONTRAIL CO<sub>2</sub> observation data assimilated between 2006 and 2010. MDM (model-data-mismatch) is a value assigned to a given site that is meant to quantify our expected ability to simulate observations and used to calculate the innovation  $X^2$  (Inn.  $X^2$ ) statistics.  $N$  denotes the number available in CTDAS. Flagged observations mean a model-minus-observation difference that exceeds 3 times the model-data-mismatch, these data are therefore excluded from assimilation. The bias is the average of the posterior residuals (assimilated values - measured values), while the modeled bias is the average of prior residuals (modeled values - measured values).

| Pressure Level | N(flagged) | MDM  | Inn.<br>$X^2$ | Bias(modeled) |
|----------------|------------|------|---------------|---------------|
| 575-625 hPa    | 0          | 2.00 | 0.00          | 0.00          |
| 485-525 hPa    | 2907(5)    | 2.00 | 0.35          | 0.05(0.08)    |
| 375-425 hPa    | 3035(3)    | 2.00 | 0.34          | -0.05(-0.07)  |
| 225-275 hPa    | 4525(4)    | 2.00 | 0.34          | 0.04(0.05)    |

**Table 3** Results of the sensitivity experiments conducted in this study (units of Pg C yr<sup>-1</sup>)<sup>a</sup>

| Inversion ID      | Case 1 | Case 2 | Case 3 | Case 4 | Case 5 | Case 6 |
|-------------------|--------|--------|--------|--------|--------|--------|
| Boreal Eurasia    | -1.02  | -0.96  | -1.11  | -1.25  | -1.03  | -0.92  |
| Temperate Eurasia | -0.68  | -0.33  | -0.70  | -0.63  | -0.37  | -0.36  |
| Tropical Asia     | 0.15   | 0.19   | 0.12   | 0.08   | 0.17   | 0.20   |
| Total Asia        | -1.56  | -1.09  | -1.69  | -1.80  | -1.23  | -1.07  |
| NH land sink      | -2.93  | -2.64  | -3.20  | -3.20  | -2.79  | -2.70  |
| Land              | -2.43  | -2.24  | -3.07  | -3.25  | -2.65  | -2.50  |
| Ocean             | -2.08  | -2.16  | -2.04  | -2.05  | -2.27  | -2.18  |
| Global            | -4.50  | -4.41  | -5.12  | -5.30  | -4.92  | -4.68  |

<sup>a</sup>The Case 1 (Surface-CONTRAIL) and Case 2 (Surface-Only) were simulated for the period 2006-2010, while Case 3-6 was simulated for the period 2008-2010; detailed discussion on global flux estimates can be found in SI Appendix B.

**Table 4** Terrestrial biosphere fluxes considered in ecosystem types for 2006-2010 (units of Pg C yr<sup>-1</sup>)

|             | type                 | Asia  | Boreal Eurasia | Temperate Eurasia | Tropical Asia |
|-------------|----------------------|-------|----------------|-------------------|---------------|
| Forest      | Total                | -0.77 | -0.71          | -0.11             | 0.04          |
|             | Conifer Forest       | -0.64 | -0.63          | -0.02             | 0.00          |
|             | Broadleaf Forest     | -0.04 | -0.01          | -0.01             | -0.01         |
|             | Mixed Forest         | -0.14 | -0.05          | -0.07             | -0.03         |
|             | Fields/Woods/Savanna | -0.01 | -0.01          | 0.00              | 0.00          |
|             | Forest/Field         | -0.02 | -0.01          | -0.01             | 0.00          |
|             | Tropical Forest      | +0.08 | 0.00           | 0.00              | +0.08         |
| Grass/Shrub | Total                | -0.44 | -0.06          | -0.36             | -0.02         |
|             | Grass/Shrub          | -0.43 | -0.06          | -0.36             | -0.02         |
|             | Scrub/Woods          | 0.00  | 0.00           | 0.00              | 0.00          |
|             | Shrub/Tree/Suc.      | 0.00  | 0.00           | 0.00              | 0.00          |
| crop        | Crops                | -0.20 | -0.02          | -0.17             | -0.01         |
|             | Total                | -0.15 | -0.23          | -0.04             | 0.13          |
|             | Semi-tundra          | -0.09 | -0.05          | -0.04             | 0.00          |
|             | Northern Taiga       | -0.17 | -0.17          | 0.00              | 0.00          |



|        |               |       |       |      |       |
|--------|---------------|-------|-------|------|-------|
| others | Wooded tundra | 0.00  | 0.00  | 0.00 | +0.06 |
|        | Mangrove      | 0.00  | 0.00  | 0.00 | 0.00  |
|        | Non-optimized | 0.00  | 0.00  | 0.00 | 0.00  |
|        | Water         | 0.07  | 0.00  | 0.00 | +0.07 |
|        | Wetland       | +0.04 | -0.01 | 0.00 | 0.00  |
|        | Deserts       | 0.00  | 0.00  | 0.00 | 0.00  |

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**Table 5** The prior/posterior land fluxes, biomass burning (fire) emissions, fossil fuel emissions and net land flux as well as the Gaussian error/their error reduction rates in Surface-Only and Surface-CONTRAIL inversion experiments during 2006-2010 (in Pg C yr<sup>-1</sup>)

| Region            | Prior Land Flux | Fire Emission | Fossil-fuel Emission | Post. Land Flux |                  | Post. Net Land Flux <sup>a</sup> |                  | Gaussian error      |
|-------------------|-----------------|---------------|----------------------|-----------------|------------------|----------------------------------|------------------|---------------------|
|                   |                 |               |                      | Surface-Only    | Surface-CONTRAIL | Surface-Only                     | Surface-CONTRAIL | Error reduction (%) |
| Boreal Eurasia    | -0.10±1.16      | 0.13          | 0.21                 | -1.09±1.05      | -1.15±0.91       | -0.96±1.05                       | -1.02±0.91       | 14                  |
| Temperate Eurasia | -0.15±0.93      | 0.03          | 3.31                 | -0.36±0.75      | -0.70±0.70       | -0.33±0.75                       | -0.68±0.70       | 6                   |
| Tropical Asia     | -0.10±0.35      | 0.32          | 0.63                 | -0.13±0.33      | -0.17±0.28       | 0.20±0.33                        | 0.15±0.28        | 15                  |
| Total Asia        | -0.35±1.53      | 0.47          | 4.15                 | -1.56±1.34      | -2.02±1.18       | -1.09±1.34                       | -1.56±1.18       | 11                  |

<sup>a</sup>Posterior Net Land Flux: including posterior land flux and fire emissions, but excluding fossil emissions.

**Table 6** Comparison of the inverted Asia terrestrial ecosystem carbon fluxes (in Pg C yr<sup>-1</sup>) from this study with previous studies

| Reference                               | Period    | Boreal Eurasia | Temperate Eurasia | Tropical Asia | Asia       | Remarks                             |
|---|-----------|----------------|-------------------|---------------|------------|-------------------------------------|
| This study                              | 2006-2010 | -1.02±0.91     | -0.68±0.70        | +0.15±0.28    | -1.56±1.18 | Surface-CONTRAIL                    |
| [Gurney <i>et al.</i> ,2003]            | 1992-1996 | -0.59±0.52     | -0.60±0.67        | +0.67±0.70    | -0.52±0.65 | -                                   |
| [Maki <i>et al.</i> ,2010]              | 2001-2007 | -1.46±0.41     | 0.96±0.59         | -0.15±0.44    | -0.65±0.49 | CNTL experiments                    |
| CTE2013 <sup>a</sup>                    | 2006-2010 | -0.93±1.15     | -0.33±0.56        | +0.22±0.20    | -1.05±1.29 | Focused on North America and Europe |
| CT2011_oi <sup>b</sup>                  | 2006-2010 | -1.00          | -0.41             | +0.14         | -1.27      | Focused on North America            |
| [Niwa <i>et al.</i> ,2012] <sup>c</sup> | 2006-2008 | -0.34±0.23     | -0.05±0.27        | +0.45±0.19    | +0.06±0.40 | GVCT                                |
|   |           | -0.25±0.28     | -0.32±0.32        | +0.03±0.29    | -0.54±0.51 | GV                                  |

<sup>a</sup>CTE2013 : Carbon Tracker Europe in Peylin et al. (2013) for the period of 2006-2010

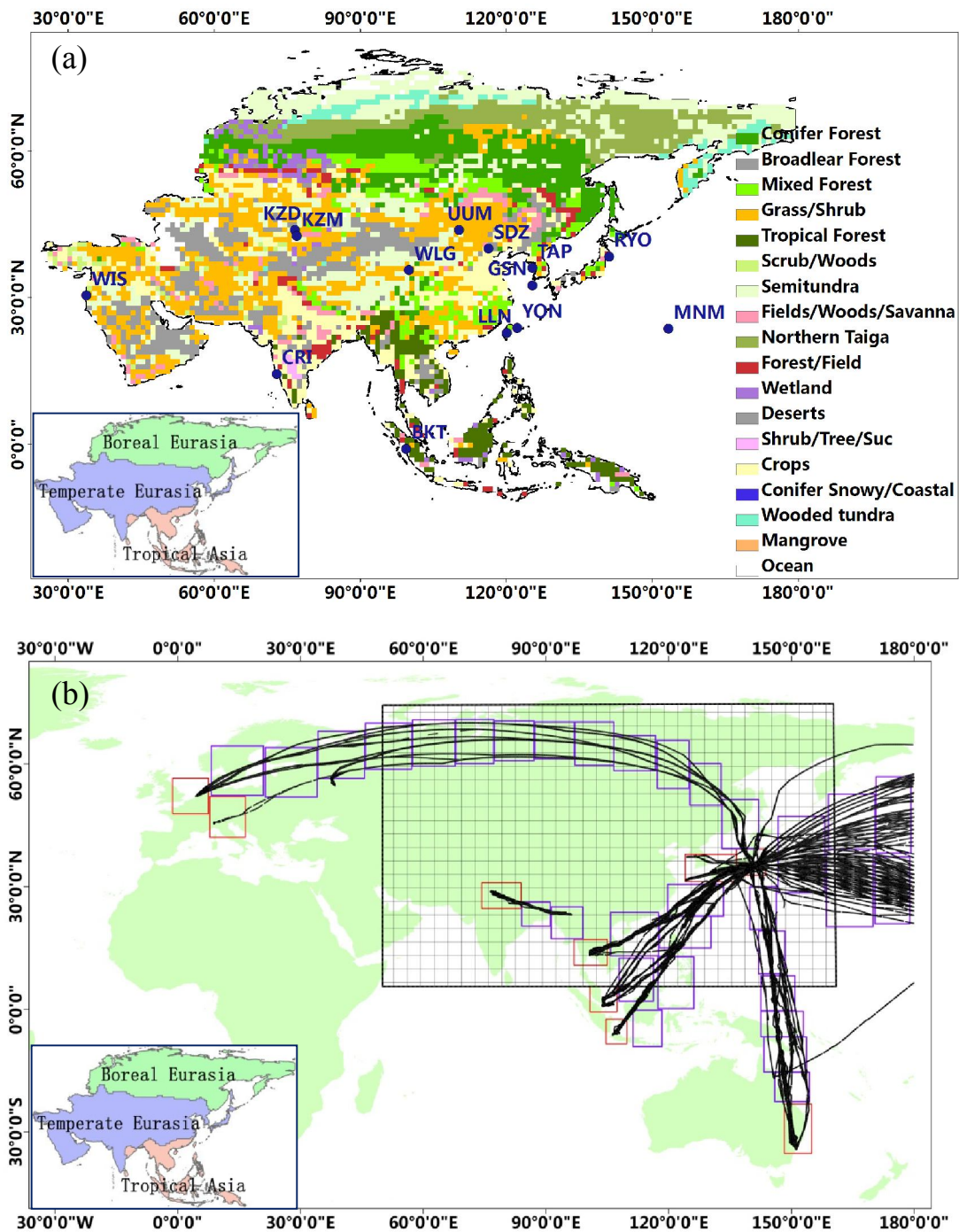
<sup>b</sup>CT2011\_oi : download from <http://carbontracker.noaa.gov>; without providing uncertainties; Note that that the CTE2013 and CT2011\_oi estimates oars not independent, and share the TM5 transport model and ObsPack observations sets, but differences in zoomed transport, state vector configuration and prior biosphere models used.

<sup>c</sup>GVCT : jointly using GLOBALVIEW and CONTRAIL CO<sub>2</sub> observation data to perform inversion ; GV : only GLOBALVIEW data used to conduct inversion ; Note that the numbers of Boreal Eurasia Temperate Eurasia and were obtain by personal communication.

**Table 7** Comparison of IAVs of the terrestrial ecosystem carbon fluxes in Asia during 2006-2010 from this study with previous studies. Fluxes (in Pg C yr<sup>-1</sup>) include biomass burning emissions but exclude fossil fuel emissions.

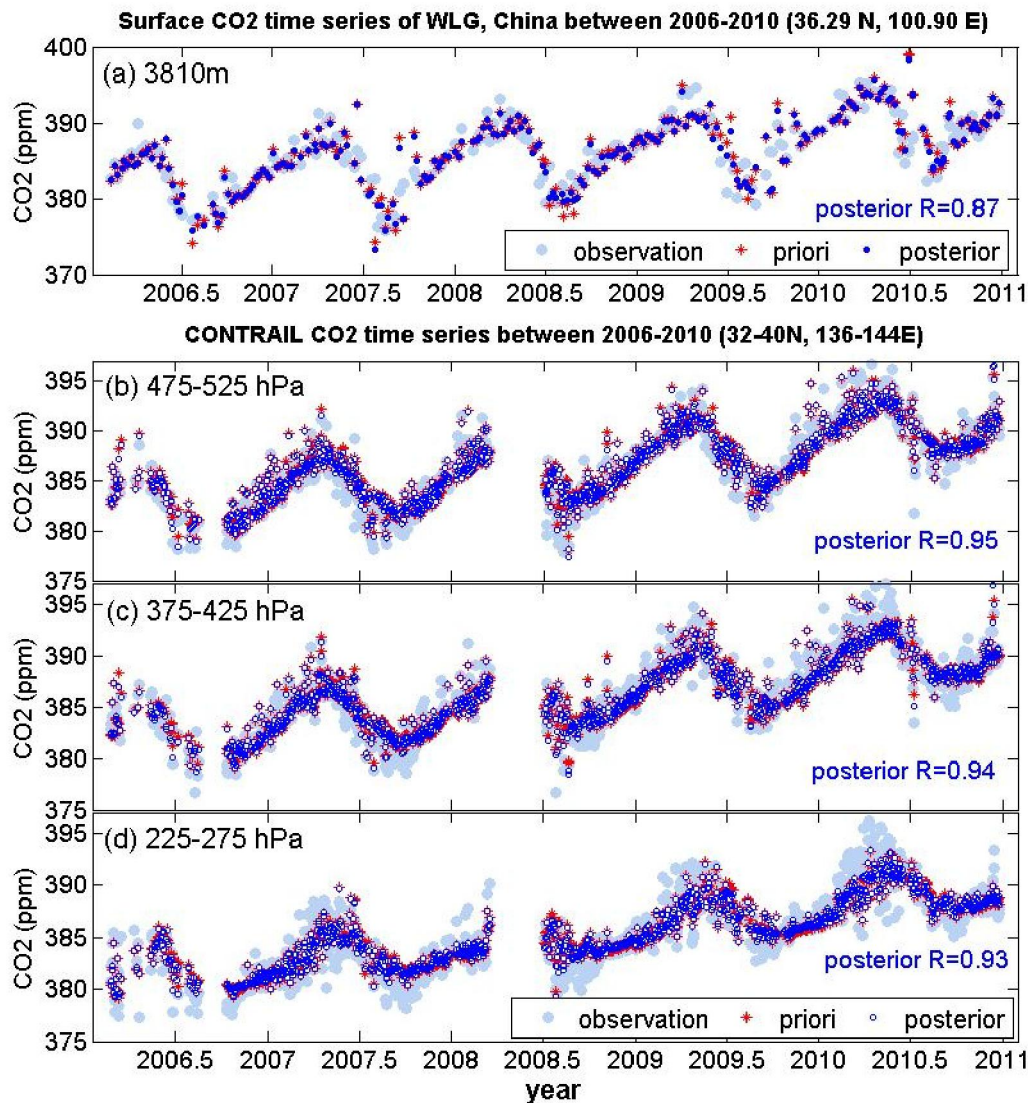
| year | Boreal Eurasia |         | Eurasia temperate |         | Tropical Asia |         |
|------|----------------|---------|-------------------|---------|---------------|---------|
|      | This study     | CTE2013 | This study        | CTE2013 | This study    | CTE2013 |
| 2006 | -0.93          | -0.93   | -0.6              | -0.4    | 0.37          | 0.41    |
| 2007 | -1.17          | -0.88   | -0.8              | -0.44   | 0.14          | 0.18    |
| 2008 | -0.96          | -1.07   | -0.66             | -0.33   | -0.09         | 0.00    |
| 2009 | -1.04          | -0.78   | -0.88             | -0.34   | 0.12          | 0.25    |
| 2010 | -1.01          | -1.02   | -0.49             | -0.12   | 0.19          | 0.27    |

Figures

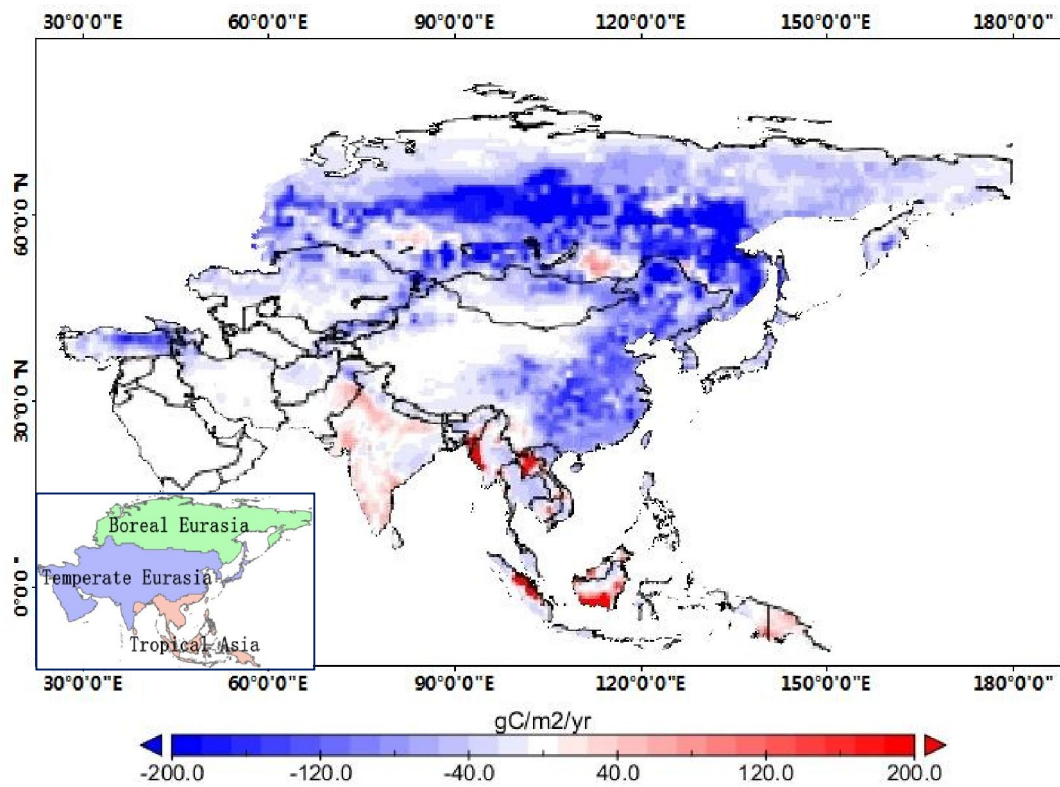


**Figure 1** (a) Map of the Asian surface observation sites, along with the map of the ecoregion types from Olson, *et al.* (1985) with 19 land cover classes as used in this study. These Asian surface observation data download from the NOAA-ESRL (e.g. WLG, BKT, WIS, KZD, KZM, TAP, UUM, CRI) and WDCGG network (e.g. LLN, SDZ, MNM, RYO, YON, GSN) ; (b) CONTRAIL CO<sub>2</sub> observations map, along with 42 horizontal regions. The red rectangles represent the 9 regions

covering the ascending and descending data (included 4 vertical bins at 575-625, 475-525, 375-425, 225-275 hPa) over airports, and the blue rectangles indicate the other 33 regions covering the cruise data (included 1 bin at 225-275 hPa). **The big black rectangle indicates a zoom region over Asia ( $1\times 1^\circ$ ) based on global grid ( $3\times 2^\circ$ ).** Note that “Asia” refers to lands as far west as the Urals in this study and it is further divided into Boreal Eurasia, Temperate Eurasia and tropical Asia based on TransCom regions (Gurney et al., 2002; Gurney et al., 2003). These divided regions are presented in the small inset in the bottom left corner (same as thereafter)

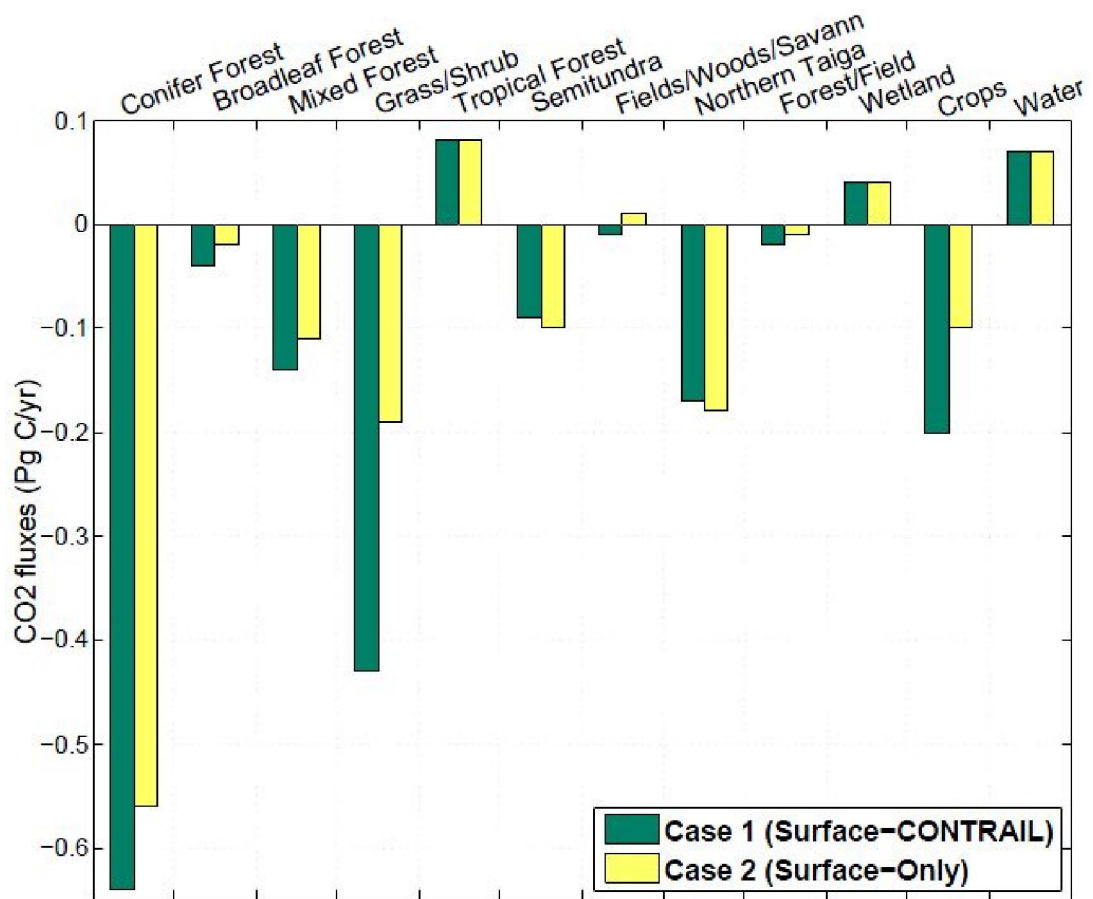


**Figure 2** Comparison of modeled values with observed CO<sub>2</sub> concentrations from surface flask station (a) Mt. Waliguan (WLG), located in China; and from CONTRAIL data in the region covering 136-144°N, 32-40°E for three different vertical bins: (b) 485-525 hPa; (c) 375-425 hPa; (d) 225-275. Although 4 vertical bins (575-625, 475-525, 375-425, 225-275 hPa) of CONTRAIL measurements have been selected and added into the system, only 3 vertical bins observations have really been assimilated as sparse measurements associated to the 575-625 hPa in CONTRAIL data. Note that the prior CO<sub>2</sub> concentrations here are not really based on a-priori fluxes only, as they are a forecast started from the CO<sub>2</sub> mixing ratio field that contains all the already optimized fluxes (1,..., n-1) that occurred before the current cycle of the data assimilation system (n). So these prior mole fractions only contain five weeks (five weeks are the lag windows in our system) of recent un-optimized fluxes and constitute our ‘first-guess’ of atmospheric CO<sub>2</sub> for each site.

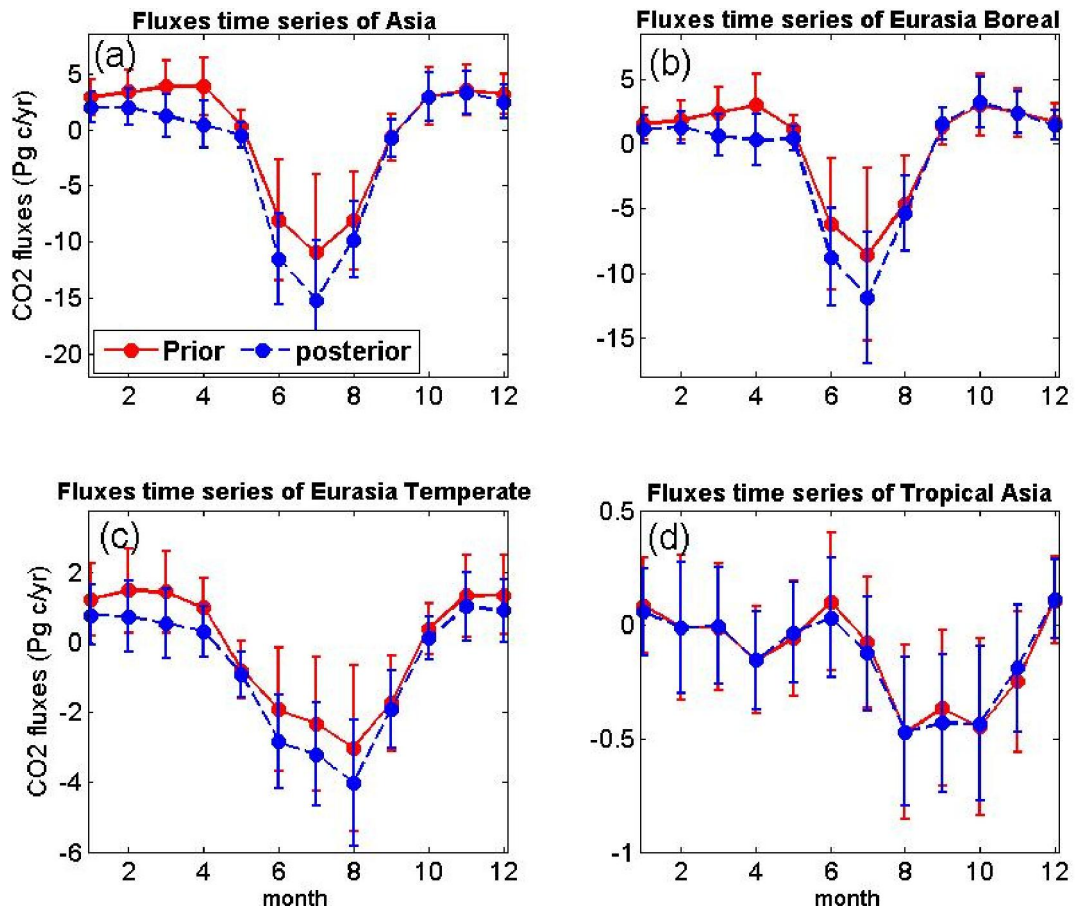


**Figure 3** Mean terrestrial biosphere carbon flux estimated from our system over Asia during 2006-2010 at a  $1 \times 1$  grid resolution. Blue colors (negative) denote net carbon uptake while red colors (positive) denote carbon release to the atmosphere. Note that the estimated flux map includes net terrestrial fluxes and biomass burning sources but excludes fossil fuel emissions.

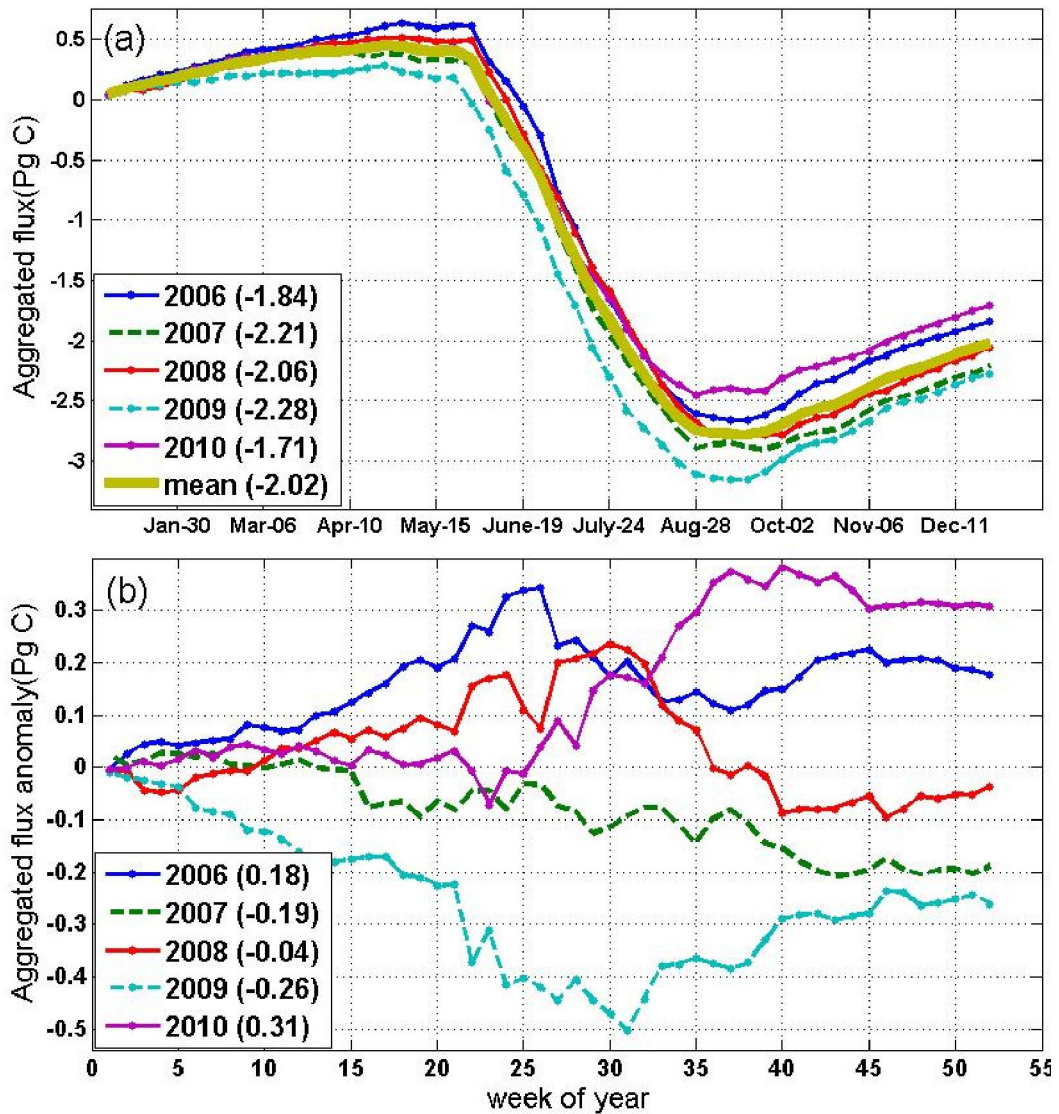




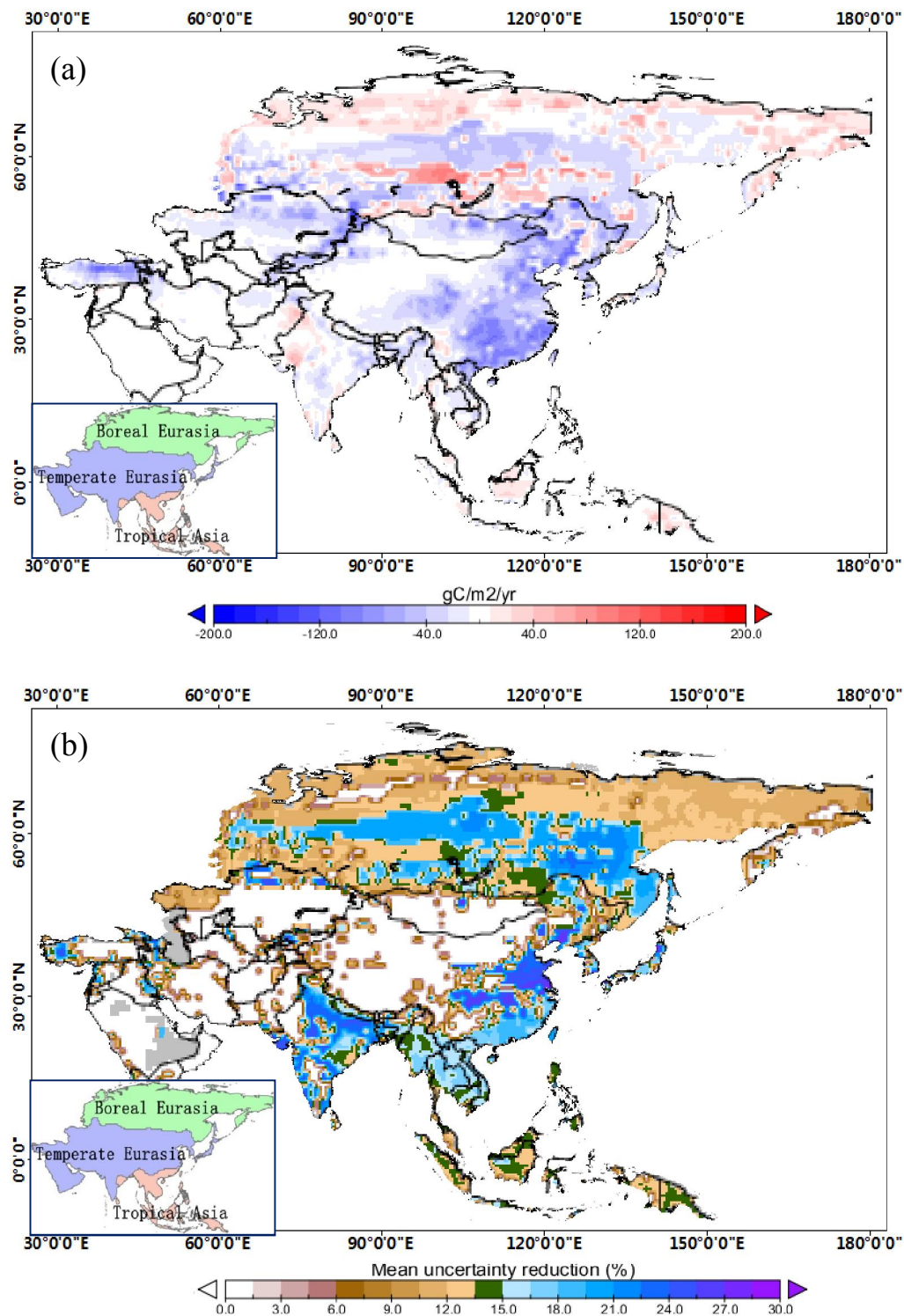
**Figure 4** Fluxes per ecoregion in Asia averaged over the period 2006-2010 in Cases 1 and 2 (in Pg C yr<sup>-1</sup>).



**Figure 5** A priori and posteriori averaged fluxes (with uncertainties) over Asian regions during 2006-2010: (a) Asia; (b) Eurasia Boreal; (c) Eurasia Temperate; (d) Tropical Asia. This flux is biosphere carbon sink after removal of fossil and biomass burning fluxes.



**Figure 6** (a) Cumulative net Asian ecosystem exchange (NEE) vs. time estimated in our system for each of the individual years and for the 2006-2010 mean. This figure reveals the largest uptake in 2009 and the smallest uptake in 2010. (b) Cumulative anomaly of Asian CO<sub>2</sub> exchange through the year 2006-2010. The inferred Asian carbon fluxes shown here include only respiration and photosynthesis, because the biomass burning emissions have a large inter-annual variability



**Figure 7** (a) The inverted flux difference between surface CO<sub>2</sub> observation data only surface (Surface-Only) and both the surface CO<sub>2</sub> observation data and CONTRAIL data (Surface-CONTRAIL); and (b) the Gaussian error reduction rate between Surface-Only and Surface-CONTRAIL during 2006-2010. The flux

difference is derived from: (Surface-CONTRAIL – Surface-Only), while the Gaussian error reduction rate is calculated as:

$$(\sigma_{Surface-Only} - \sigma_{Surface-CONTRAIL}) / \sigma_{Surface-Only} \times 100$$