

Carbon balance of China constrained by CONTRAIL aircraft CO₂ measurements

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Abstract: Terrestrial CO₂ flux estimates in China using atmospheric inversion method are beset with considerable uncertainties because very few atmospheric CO₂ concentration measurements are available. In order to improve these estimates, nested atmospheric CO₂ inversion during 2002 - 2008 is performed in this study using passenger aircraft-based CO₂ measurements over Eurasia from the Comprehensive Observation Network for Trace gases by Airliner (CONTRAIL) project. The inversion system includes 43 regions with a focus on China, and is based on the Bayesian synthesis approach and the TM5 transport model. The terrestrial ecosystem carbon flux modeled by the BEPS model and the ocean exchange simulated by the OPA-PISCES-T model are considered as the prior fluxes. The impacts of CONTRAIL CO₂ data on inverted China terrestrial carbon fluxes are quantified, the improvement of the inverted fluxes after adding CONTRAIL CO₂ data are rationed against climate factors and evaluated by comparing the simulated atmospheric CO₂ concentrations with three independent surface CO₂ measurements in China. Results show that with the addition of CONTRAIL CO₂ data, the inverted carbon sink in China increases while those in South and Southeast Asia decrease. Meanwhile, the posterior uncertainties over these regions are all reduced (2~12%). CONTRAIL CO₂ data also have a large effect on the inter-annual variation of carbon sinks in China, leading to a better correlation between the carbon sink and the annual mean climate factors. Evaluations against the CO₂ measurements at three sites in China also show that the CONTRAIL CO₂ measurements may have improved the inversion

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30 results.

31

32 **1. Introduction**

33 Carbon dioxide (CO₂) and other greenhouse gases emitted from human activities are the main
34 cause of global warming (IPCC, 2007). Terrestrial ecosystems play a very important role on
35 regulating the atmospheric CO₂ concentration. According to the evidence from atmosphere
36 observations, Le Quéré et al. (2009) estimated that the mean CO₂ uptake rate of the global
37 terrestrial ecosystem is 2.6±0.7 Pg C yr⁻¹ for 1990–2000, which offsets 40% of the global
38 fossil fuel carbon emissions. Pacala et al. (2001) found that North American terrestrial
39 ecosystems sequestered 30% - 50% of its industrial CO₂ emissions in the 1980s. China has a
40 vast land area of 960×10⁶ ha, and nearly 80% of the land areas are covered with various types
41 of vegetation, including forest (16.5%), grass (34.8%), shrubs (18.5%), croplands (11.2%),
42 and other types (Fang et al., 2007). Using the methods of bottom–up (inventory survey and
43 process-based ecosystem model) and top-down (atmospheric inversion) approaches, many
44 studies have been conducted during the past decade to estimate China’s terrestrial ecosystem
45 carbon sinks (e.g., Cao et al., 2003; Fang et al., 2007; Piao et al., 2009; Tian et al., 2011;
46 Jiang et al., 2013). Fang et al. (2007) estimated the land sinks in China could offset 20.0 –
47 26.8% of its industrial carbon emissions. However, there are still large gaps of land sink
48 derived using bottom-up and top-down methods (Piao et al., 2009). **One of the main reasons**
49 **may be attributed to the lack of enough CO₂ concentration observations. Jiang et al. (2013)**
50 **pointed out that due to lack of sufficient observations, most regions of the country have a low**
51 **regional and annual uncertainty reduction percentage (< 10%) by way of atmospheric**
52 **inversion, especially for South and Southwest China, and the overall uncertainty reduction is**
53 **relatively lower compared with North America and Europe.**

54 The lack of surface measurements can partially be compensated for by aircraft
55 measurements in the free troposphere. Many vertical profiles of CO₂ have been obtained over
56 Europe and North America using research aircraft (Crevoisier et al., 2010; Xueref-Remy et al.,
57 2011). Compared with research aircrafts, passenger aircraft CO₂ measurements are done at
58 much lower cost, and could cover larger areas. Presently, there are two well-known CO₂
59 measurement projects using passenger aircrafts, the Civil Aircraft for the Regular

60 Investigation of the atmosphere Based on an Instrument Container (CARIBIC) project
61 (Brenninkmeijer et al., 2007; Schuck et al., 2009) and the Comprehensive Observation
62 Network for Trace gases by Airliner (CONTRAIL) project (Machida et al., 2008; Matsueda et
63 al., 2008). CARIBIC measures atmospheric CO₂ by flask sampling between Germany and
64 destinations in Europe, Africa, North and South America and Asia four flights per month,
65 while CONTRAIL measures CO₂ continuously between Japan and Europe, Australia, South
66 and Southeast Asia, and North America. Patra et al. (2011) performed an inversion study
67 based on CARIBIC data and evaluated the results against CONTRAIL data. With the
68 CONTRAIL measurements, Niwa et al. (2012) conducted an inverse modeling study with a
69 focus on tropical terrestrial regions. Their results showed that CONTRAIL data have a large
70 impact on the inversion results.

71 In this study, a China-focused atmospheric inversion is conducted using CONTRAIL
72 measurements over Eurasia. The inversion system and CONTRAIL data used are first
73 described, followed by presentation and discussion of the impact of CONTRAIL data on
74 inverted carbon fluxes and posterior uncertainties in China and its surrounding areas.

75 **2. Method and Data**

76 **2.1 Inversion setting**

77 In this study, a nested inversion system (Jiang et al., 2013) based on the Bayesian synthesis
78 inversion method (Rayner et al., 1999; Enting et al., 1989) is used to improve the estimations
79 of **monthly** CO₂ sources and sinks as well as their uncertainties. The global surface is
80 separated into 43 regions based on the 22 TransCom large regions (e.g. Gurney et al., 2003;
81 Baker et al., 2006), with 13 small regions in China (Figure 2). The partition scheme for the 13
82 small regions is mainly based on land cover types, i.e. forest (5 regions), crop (4 regions),
83 grass (3 regions), and desert (1 region).

84 A monthly transport operator for the 43 regions and forward transport simulations for
85 carbon emissions from fossil fuel and biomass burning are calculated using a global two-way
86 nested transport model TM5 (Krol et al., 2005). The fossil fuel inventory is from the Miller
87 Carbon Tracker fossil fuel emission field (CarbonTracker, 2010), which is constructed based
88 on CDIAC 2007 (Boden et al., 2010) and EDGAR 4 databases (Olivier and Berdowski, 2001).

89 The biomass-burning inventory is from the Global Fire Emissions Database version 3
90 (GFEDv3) (van der Werf et al., 2010). Except for sources and sinks, the key processes of CO₂
91 in the atmosphere are horizontal and vertical transport and diffusion. In the TM5 model, the
92 horizontal transport is based on the slopes advection scheme (Russel and Lerner, 1981), the
93 convection is parameterized according to Tiedtke (1989), and the vertical diffusion near
94 surface layer and in the free troposphere are parameterized using the schemes of Holtslag and
95 Moeng (1991) and Louis (1979), respectively. An evaluation showed that TM5 has very well
96 performs on vertical and horizontal transport (Stephens et al., 2007). In this study, TM5
97 model is driven by the ECMWF outputs, and is run at a horizontal resolution of 3° × 2°
98 around the world without nesting a high-resolution domain and a vertical structure of 25
99 layers, with the model top at about 1 hPa.

100 Hourly terrestrial ecosystem carbon exchanges simulated by the BEPS model (Chen et
101 al., 1999; Ju et al., 2006) and daily carbon fluxes across the air-water interface calculated by
102 the OPA-PISCES-T model (Buitenhuis et al., 2006) are considered as prior fluxes. Before
103 being used in the inversion, both these two type of fluxes are averaged to monthly. In addition,
104 the monthly terrestrial carbon fluxes of each grid are neutralized annually. The 1σ
105 uncertainties for the prior fluxes over the global land and ocean surfaces are assumed to be
106 2.0 and 0.67 PgC yr⁻¹, respectively, which are the same as those used in Deng and Chen
107 (2011). That is because we use the same land and ocean prior fluxes with Deng and Chen
108 (2011). Deng and Chen (2011) did χ^2 test for their selections, and results indicated that these
109 uncertainties are reasonable. The uncertainty on the land is spatially distributed based on the
110 annual NPP distribution simulated by BEPS, while the one on the ocean is distributed
111 according to the area of each ocean region. The monthly uncertainties for the terrestrial
112 regions are assigned according to the variations of monthly NPP, while the ones for oceanic
113 regions are assumed to be even. We do not consider the relationship among different regions.
114 Hence, a diagonal matrix for error variances is used. That is because the global land is
115 separated into a series of regions mainly according to land cover types, and we assume that
116 the relationship of the fluxes of different land cover types could be negligible.

117 A total of 130 sites of CO₂ observations from GLOBALVIEW-CO₂ 2010 dataset

118 (GLOBALVIEW-CO₂, 2010) are adopted, including 54 flask observations, 7 continuous
119 measurements, 5 tower sites, 6 ship sites, and 58 aircraft sites. The locations of the
120 GLOBALVIEW (GV) CO₂ observations could be found in Figure 2. The model-data
121 mismatch error in ppm is defined using the following function, which is similar to those used
122 by Peters et al. (2005) and Deng and Chen (2011).

$$123 \quad R = \sigma_{const}^2 + GVsd^2$$

124 where *GVsd* reflects the observation error, it is the standard deviation of the residual
125 distribution in the average monthly variability (var) file of GLOBALVIEW-CO₂ 2010. The
126 constant portion σ_{const} reflects the simulation error, which varies with station type that is
127 because the transport models generally have different performances at different observation
128 stations. Except for some difficult stations, the observation sites are divided into 5 categories.
129 The categories (respective value in ppm) are: Antarctic sites/oceanic flask and continuous
130 sites (0.30), ship and tower sites (1.0), mountain sites (1.5), aircraft samples (0.5), and land
131 flask/continuous sites (0.75). The value of 3.5 is used for the difficult sites (e.g., abp_01D0,
132 bkt_01D0).

133 **2.2 CONTRAIL aircraft CO₂ measurements**

134 The CONTRAIL project measures CO₂ concentrations using continuous measurement
135 equipment on passenger aircrafts (Machida et al., 2008; Matsueda et al., 2008). Calibrations
136 and evaluations have shown that the accuracy of CONTRAIL data compares well with the
137 GV CO₂ data (Machida et al., 2011). In this study, aircraft CO₂ measurements over Eurasia
138 during November 2005 to December 2009 are used. The measurements between Japan and
139 Europe, Korea, Taiwan, South Asia and Southeast Asia are shown in Figure 1a. In order to
140 use the continuous observations in the inversion system, the measurements are divided into 87
141 sites, including 19 level flight sites (~10 km, Figure 1a) and 68 vertical sites. Following Niwa
142 et al. (2012), each vertical measurement profile is divided into 5 layers: 0 ~ 1000m,
143 1000~2000m, 2000 ~4000m, 4000~6000m, and 6000~8000m. The flight altitude higher than
144 8000 m is considered as level flight. For the level flight sites, first, we define 19 regions with
145 each of size 10 × 10 degrees according to the flight routes (Figure 1a); then, all the
146 observation records with altitude higher than 8000 m located in one region are averaged to get

147 the concentration of that site, and its location is determined by averaging the location of all
148 observation records as well. It should be noted that the $10^\circ \times 10^\circ$ boxes are only applied to the
149 level flight. Before being used in the inversion, 1) daily mean data for each site are smoothed
150 (Figure 1b) using the same technique as Masarie and Tans (1995), and then averaged to
151 monthly mean value; 2) the sites with samples shorter than 6 months are excluded; 3) the
152 variations of the monthly CO₂ concentrations for each site are carefully checked, and we find
153 that in the boundary layer (below 2000 m), the concentrations at most sites are highly affected
154 by local emissions, probably emitted by frequent aircraft ascending and descending, hence, only
155 data measured above 2000 m are used. Niwa et al. (2012) also only used the free tropospheric
156 (above 625 hpa) data. In addition, in previous studies (Sawa et al., 2008; Niwa et al. 2012),
157 the CO₂ recodes in the stratosphere were filtered out, because the seasonal variation of CO₂ in
158 stratosphere is quite different from that in troposphere. However, in this study, we don't
159 distinguish whether the CO₂ recodes are in stratosphere or in troposphere. We have checked
160 the monthly observed and forward simulated CO₂ concentrations at the level flight sites and
161 thought that the influences of stratospheric CO₂ could be neglectful in our inversion system.
162 Consequently, 54 CONTRAIL CO₂ sites are used in the inversion (Figure 2). The model-data
163 mismatch errors of the CONTRAIL data are calculated using the same method as those of the GV
164 data. The observation error (GV_{sd}) is the standard deviation of the residual after smoothing, and
165 for the item of σ_{const} , considering the large range ($10^\circ \times 10^\circ$) of the level flight sites and the
166 thick layer of the vertical sites, we use a constant of 0.75 ppm, which is larger than that of GV
167 aircraft samples (0.5 ppm).

168

169 **2.3 CO₂ measurements in China**

170 In this study, CO₂ concentrations measured during Jul 2006 – Dec 2009 at three Chinese sites
171 are used to evaluate forward simulation results. The three sites were all established by
172 Chinese Academy of Meteorological Sciences (CAMS), China Meteorological Adminis-
173 tration (CMA), with names of Longfengshan (LFS), Shangdianzi (SDZ) and LinAn (LAN),
174 respectively. LFS, SDZ and LAN are located in Northeast China, North China, and East
175 China, respectively (Figure 3). The weekly flask measurements of these stations are sampled

176 and analyzed using the recommended methods of the Global Atmosphere Watch programme
177 of World Meteorological Organization (WMO/GAW), and the results are comparable to that
178 of the Earth System Research Laboratory in National Oceanic and Atmospheric
179 Administration (NOAA/ESRL). For more detailed information about these observations,
180 please refer to Liu et al. (2009).

181 **3. Results and discussions**

182 **3.1 Impact on inter-annual variations**

183 Two inversion experiments are conducted from 2000 to 2009, the first (Case GV) run with
184 only GLOBALVIEW CO₂ included, the second run with CONTRAIL CO₂ data added (Case
185 GVCT). For each experiment, the first two years are considered to be the spin-up period, and
186 the last year is the spin-down time. Therefore, the analysis period in this study is from 2002 to
187 2008.

188 Figure 4 shows the inter-annual variations (IAVs) of the inverted land sinks (excluding
189 biomass burning emissions, the same thereafter) and the posterior uncertainties of the two
190 experiments in China. When only GV CO₂ is used, the IAVs show a less negative trend, with
191 strongest land sink occurring in 2002, and weakest sink in 2008. When CONTRAIL CO₂ is
192 added after December 2005, changes of inverted sinks occur after 2005, with largest change
193 in 2007, compared to Case GV. **The CONTRAIL CO₂ also has an impact on the posterior
194 uncertainties from 2005 to 2008, especially in 2007 and 2008. The posterior uncertainties in
195 2007 and 2008 are reduced from around 0.195 to 0.183 PgC yr⁻¹, with a reduction rate of 6%.
196 This reduction rate is defined as $(Uncertainty_{posterior,GVCT} - Uncertainty_{posterior,GV}) \times 100 /$
197 $Uncertainty_{posterior,GV}$ (same thereafter). In our another study (Jiang et al., 2013), we added CO₂
198 measurements of the three Chinese sites in the same inversion system, and results showed that the
199 posterior uncertainties were reduced from around 0.195 to 0.173 PgC yr⁻¹ (~11%) in 2007 and
200 2008. Hence, We think that this uncertainty reduction caused by adding the CONTRAIL data is
201 reasonable in comparison with the uncertainty reduction caused by adding the three Chinese sites
202 and in consideration of the fact that there are no CONTRAIL data in China: all data were
203 measured in the downwind or upwind of China and only data above the boundary layer are used in
204 the inversion.** Overall, when the CONTRAIL CO₂ data are added, the inverted carbon sink in

205 China increases by 0.05 PgC yr^{-1} (30%), the posterior uncertainty is reduced by 0.0043 PgC
206 yr^{-1} (2.2%), and the mean carbon sink in China is $-0.25 \pm 0.19 \text{ PgC yr}^{-1}$ for 2002 to 2008.

207 Climate factors such as temperature, precipitation and radiation could affect plant growth
208 (Zhu et al., 2007; Myoung et al., 2013), thereby the IAVs of land sinks (Ciais et al., 2005).
209 Generally, a warmer condition advances vegetation growth for most regions in middle and
210 high latitudes of the Northern Hemisphere (NH), including the crops in Europe and US, and
211 the forests in central Siberia, west Canada and northeast China (Myoung et al., 2013). Zhu et
212 al. (2007) also showed that in northern China, the plant growth was temperature-limited.
213 Usually, more vigorous vegetation growth corresponds to more carbon uptake. Studies on the
214 relationships between the net ecosystem exchange (NEE) measured by eddy covariance
215 equipment and the environmental factors confirm that the IAVs of annual mean air
216 temperature is significantly related to the ones of NEE in the forest regions in middle and
217 high latitudes of the NH: Yuan et al. (2009) pointed that air temperature was the primary
218 environmental factor that determined the IAV of NEE in deciduous broadleaf forest across the
219 North American sites, and NEE was positively correlated with the mean annual air
220 temperature; Dunn et al. (2007) found that in central Manitoba, Canada, warmer annual
221 temperatures were associated with increased net uptake, while annual precipitation did not
222 explain any of the variability in NEE. Nevertheless, in low latitudes of the NH, for example,
223 southern China, the NEE may be related to solar radiation and precipitation. Zhu et al. (2007)
224 reported that in southern China, the plant growth was radiation-limited. Usually, more
225 radiation corresponds to less cloud cover, so as to less precipitation (Figure 5a). Based on
226 eddy covariance measurements in two forest sites in southern China, Yan et al. (2013) found
227 that the greater annual NEE (more uptake) usually occurred in the dry years and smaller
228 annual NEE in the rainy years for the both forests. Hence, in order to study whether the
229 impact of CONTRAIL data is reasonable, it should be useful to check the relationship
230 between inter-annual variations of climate factors and land sinks in different regions of China.
231 Because the changes of posterior fluxes mainly occur in southern China and northern China
232 (see section 3.2), only these two regions are investigated. Monthly climate data of 484
233 stations obtained from CMA during the study period are used for the analysis, in which 269
234 stations are located in southern China, and the others are located in northern China (Figure 3).

235 Figure 5 shows the IAVs of annual mean climate factors anomalies and land sink anomalies
236 in southern China and northern China. It could be found that in southern China, the changes
237 caused by CONTRAIL data lead to a better correlation between the fluxes and radiation as
238 well as precipitation, and in northern China, the changes also make the fluxes better
239 correlated with the temperature. These relationships are consistent with the previous findings,
240 indicating that the IAVs of posterior fluxes in China by additional constraint of CONTRAIL
241 CO₂ data could be more reasonable.

242 3.2 Impact on spatial pattern

243 As shown in section 3.1, CONTRAIL CO₂ has a large impact on the inverted carbon
244 fluxes and a certain impact on the uncertainties during the 2006 – 2008 period. Furthermore,
245 the impacts of CONTRAIL CO₂ on the spatial patterns of the inverted global carbon fluxes
246 and uncertainties for 2006 to 2008 are shown in Figure 6. For Case GV, most of the land
247 regions are found to be carbon sinks (Figure 6a), with strong sinks ($> 50 \text{ gC m}^{-2}\text{yr}^{-1}$) occurring
248 in Boreal Asia, South and Southeast Asia, the eastern U.S. and southern South America (S.A.),
249 while Tropical America and Southern Africa appear as carbon sources. Most China regions
250 appear as weak carbon sinks ($< 30 \text{ gC m}^{-2}\text{yr}^{-1}$). Comparing Case GVCT with the Case GV, the
251 carbon sinks decrease in South and Southeast Asia, tropical Africa, boreal and western
252 temperate North America, and Southwest China, with the most significant decrease happening
253 in Southeast Asia ($> 50 \text{ gC m}^{-2}\text{yr}^{-1}$). The carbon sinks increase in Europe, boreal and western
254 Asia, eastern temperate North America, eastern China, southern Africa and most of the
255 Pacific, with the most notable increase in eastern China. It should be noted that though only
256 CONTRAIL CO₂ data over Eurasia are used in this study, its impacts are global. Meanwhile,
257 posterior uncertainties over most of the Northern Hemisphere and tropical regions are reduced,
258 with the most significant reduction occurring in South and Southeast Asia ($\sim 10\%$). In China,
259 the uncertainty reduction in all regions is smaller than 5 %, with largest reductions in East and
260 Southwest China.

261 Compared with results from Niwa et al. (2012) for the same time period, the decrease of
262 land sink in Southeast Asia, increases of land sinks in Europe, boreal Asia, eastern temperate
263 North America, and southern Africa, and the reductions of posterior uncertainties in South
264 and Southeast Asia are consistent. However, in South Asia and eastern China, although large

265 effects of CONTRAIL CO₂ data on land sinks were also derived in Niwa et al. (2012), the
266 impacts are opposite. Moreover, the magnitude of the effects on the inverted land fluxes and
267 associated uncertainties are much smaller than those in Niwa et al. (2012). In order to gain
268 insight on the causes of the differences between this study and Niwa et al. (2012), mean
269 monthly prior and posterior fluxes of Case GV and Case GVCT in China, South Asia and
270 Southeast Asia (including Indo-China Peninsula and tropical Asia) during 2006 – 2008 are
271 compared in Figure 7. In China, there are large differences in the prior fluxes. The seasonal
272 amplitude of Niwa's fluxes (simulated using CASA model) is much larger than that
273 (simulated using BEPS model) of this study. After being constrained by GV and CONTRAIL
274 CO₂ data, the posterior fluxes from this study and Niwa et al. (2012) tend to be close to each
275 other: the land sinks during growing season increase in this study, while those decrease in
276 Niwa et al. (2012), and the strongest sinks are very similar in magnitude. However, the flux
277 estimated in this study turns from source to sink in May, reaching the strongest in July, while
278 that estimated in Niwa et al. (2012) turns from source to sink in July, reaching the strongest in
279 August. This delay leads to a relatively large divergence in the inverted annual carbon sink
280 (-0.29 versus 0.25 PgC yr⁻¹). In contrast, in South Asia, the seasonal amplitude of Niwa's
281 prior fluxes is smaller than that of this study. When only being constrained with GV CO₂ data,
282 the land sinks during the growing season obviously increase in this study, while little changes
283 happen with Niwa's results. After CONTRAIL CO₂ data are added, the sinks during the
284 growing season decrease, and the sources during non-growing season increase in this study,
285 leading to a decrease in the annual land sink; while in Niwa et al. (2012), the land sources
286 increase significantly during the non-growing season, and the land sinks during the growing
287 increase remarkably as well. The annual land sinks in South Asia from these two studies are
288 very close. In Southeast Asia, there are also significant differences in the prior fluxes:
289 compared with Niwa's results, there are much higher carbon sinks from May to Oct, and
290 higher sources from Dec to Apr in this study. When only being constrained with GV CO₂ data,
291 except in Oct and Nov, more carbon is absorbed in all month in this study, especially in Feb
292 and Sep; while in Niwa et al. (2012), the land sinks increase in Mar, Sep and Oct, and in the
293 other months, the land sources increase. The annual land sink increases from 0.0 PgC yr⁻¹ to
294 -0.68 PgC yr⁻¹ in this study, while little changes occur in Niwa et al. (2012). This difference

295 may partly due to the use of Bukit Koto Tabang (BKT) station in Indonesia and the
296 CONTRAIL-ASE observations between Australia and Japan included in GLOBALVIEW
297 dataset in this study (Figure 2), which were not used in Niwa et al. (2012). After the
298 CONTRAIL CO₂ data are added, the carbon sinks decrease in all months in this study, and
299 they decrease in most months in Niwa et al. (2012) as well. The monthly variations of the
300 posterior fluxes between this study and Niwa et al. (2012) are similar to a certain extent,
301 especially from Sep to Dec. However, from Jan to Aug, the carbon sources in Niwa et al.
302 (2012) are significantly higher than that of this study, especially in Jan, Feb, Jun and Jul.
303 Overall, the uses of different prior fluxes and amount of GV observations may result in these
304 different effects of CONTRAIL CO₂ data. However, the posterior fluxes from these two
305 studies tend to be close to each other after being constrained with CONTRAIL CO₂ data.

306 The statistics show that with the further constraint of CONTRAIL CO₂ data, the carbon
307 sink in China increases from -0.16 ± 0.19 PgC yr⁻¹ to -0.29 ± 0.18 PgC yr⁻¹. At the same time,
308 the land sinks of Southeast Asia and South Asia decrease from -0.68 ± 0.34 and -0.28 ± 0.32
309 PgC yr⁻¹ to -0.35 ± 0.30 and -0.11 ± 0.30 PgC yr⁻¹, respectively. When CONTRAIL data are
310 added, the land sink in China is close to the inversion result of -0.35 PgC yr⁻¹ stated in Jiang
311 et al. (2013) for the same period, which was derived by adding three additional China CO₂
312 observation stations. The land sink obtained for South Asia, -0.11 ± 0.30 PgC yr⁻¹, agrees well
313 with the -0.104 ± 0.15 PgC yr⁻¹ result of Patra et al., (2013) for 2007-2008 period. Southeast
314 Asia is one of the most forested regions in the world. Its land sink should be dominated by
315 forest. Pan et al. (2011) estimated that here the forest carbon flux from 2000 to 2007 was
316 -0.12 PgC yr⁻¹. Carbon emission from biomass burning is 0.30 PgC yr⁻¹ in this region from
317 GFEDv3, so, the net carbon flux, excluding fossil fuel emissions, is -0.05 PgC
318 yr⁻¹ ($-0.35+0.30=-0.05$ PgC yr⁻¹), which is comparable to the results of Pan et al. (2011). The
319 main reason of the significant changes in South and Southeast Asia is that there are very few
320 CO₂ measurements in the GV dataset in these regions (Figure 2), so there is an insufficient
321 observational constraint, leading to large uncertainties in the inverted carbon fluxes. The
322 addition of CONTRAIL data reduces uncertainty by markedly increasing observations in
323 these regions. Despite the fact that most CONTRAIL CO₂ measurements are in the middle
324 and upper troposphere, due to strong tropical convection, the resulting observational

325 constraints are still relatively strong, thus decreasing land sinks in these regions. Niwa et al.
326 (2012) have detailed the mechanisms of CONTRAIL CO₂ data impacts on the fluxes. The
327 large changes in Eastern China could be explained with that there are many CONTRAIL CO₂
328 measurements over East China Sea, Korea and Japan, which are mostly downwind of China,
329 so CONTRAIL CO₂ over these regions could directly sense carbon fluxes in eastern China to
330 a certain extent. Figure 8 shows contributions from emissions in different regions of the globe
331 at a current month (July 2007) and for the previous five months (1 Pg carbon emitted from
332 each region at one month) to the CO₂ concentration in July 2007 at 2000 – 4000 m height
333 over Taipei airport (TPE). There are strong contributions from the emissions of eastern China
334 (South China, East China forest, and Yangtze plain) at the current month (July 2007).
335 Therefore, CONTRAIL CO₂ data could affect the inversion results in China.

336 **3.3 Evaluation against CO₂ measurements in China**

337 We further use the CO₂ concentration measured at the three Chinese observation sites, i.e.,
338 LFS, SDZ, and LAN, to evaluate the forward simulation results using the TM5 model from
339 the posterior fluxes. The weekly measurements are smoothed and extrapolated to obtain
340 monthly values, using the same technique as GV CO₂ dataset (Masarie and Tans, 1995). The
341 simulations are conducted from 2000 to 2009, with initial concentration of 368.75 ppm (Ed
342 Dlugokencky and Pieter Tans, NOAA/ESRL, www.esrl.noaa.gov/gmd/ccgg/trends/). Since
343 the impact of CONTRAIL CO₂ is the largest in 2007, the simulation results in 2007 are
344 evaluated using the CO₂ concentration measurements (Figure 9). Obviously, the simulated
345 CO₂ concentrations using the posterior fluxes constrained by CONTRAIL data are much
346 closer to the observations during the summertime at all three sites. The mean biases between
347 the simulations and observations at LAN, LFS and SDZ are reduced from 2.13, 4.39, and 3.62
348 ppm to 1.28, 3.40, and 2.74 ppm, respectively. Moreover, the seasonal amplitude of CO₂
349 concentration between wintertime and summertime may better reflect the seasonal variations
350 in land ecosystem sources and sinks in the upwind areas of the stations. Except at SDZ,
351 simulated concentration amplitude using posterior fluxes constrained by CONTRAIL data is
352 also much closer to the observations (Figure 9) than those without the CONTRAIL constraint.
353 Therefore, CONTRAIL data may have helped improve the inversion results for China at a
354 certain extent.

355 **3.4 Sensitivity analysis**

356 It could be found that the changes caused by CONTRAIL data over China, including the
357 inverted monthly carbon sinks (Figure 7a) and the concentrations simulated using the inverted
358 fluxes (Figure 9), mainly occur in the warm seasons. This phenomenon may be attributed to
359 the inversion setup for the monthly prior fluxes uncertainties, i.e., the monthly prior errors for
360 the terrestrial regions are assigned according to the variations of monthly NPP (Section 2.1),
361 which lead to the prior errors in the cold seasons are very small, especially in the high latitude
362 areas. Therefore, we conduct two sensitivity experiments, namely Case GV_s and Case
363 GVCT_s, in which both variations of monthly NPP and soil respiration (RESP) are
364 considered when we assign the month prior errors.

365 As shown in Figure 10a, in China, after both considered the NPP and RESP, the monthly
366 prior uncertainties in summer are reduced, while those in cold seasons are enhanced,
367 especially in northern China. These changes cause the land sinks decrease in Northeast China
368 and West China and increase in the other regions of China (not shown). As an aggregate, in
369 southern China, the land sink increases in most years, but the IAVs are the same as before; in
370 northern China, basically, there is no change. Seasonally, in southern China, the land sinks
371 increase in all months, and in northern China, they increase in spring and decrease in autumn.
372 As a whole in China, though the land sink decreases in 2004 and increases in 2007 at a certain
373 level, and little changes occur in the other years, but the IAVs are basically the same as before.
374 Seasonally, more carbon uptake occurs in spring and less happens in autumn (Figure 10b, c),
375 leading to the simulated concentrations decrease in spring and more closer with the
376 observations at all three sites (Figure 10d). However, the gaps between the simulated and the
377 observed concentrations during the cold seasons are still large, especially at LAN and SDZ.
378 Since LAN is close to Hangzhou City, and SDZ is close to Beijing City, these large gaps may
379 be related to the model resolution. Overall, the mean land sinks during 2002-2008 in China
380 are 0.187 ± 0.20 and 0.255 ± 0.20 PgC yr⁻¹ for Case GV_s and Case GVCT_s, respectively,
381 which are almost the same as the values of -0.194 ± 0.19 and -0.253 ± 0.19 PgC yr⁻¹ for Case
382 GV and Case GVCT, indicating that the different settings of monthly prior errors have little
383 impact on the inverted carbon budget in China.

384 **4. Summary and Conclusions**

385 In this study, CONTRAIL Aircraft CO₂ measurements over Eurasia are used to constrain the
386 inversion besides GV CO₂ data in a nested atmospheric inversion system with the focus on
387 China during 2002 -2008. The CONTRAIL CO₂ measurements are grouped into 87 sites, and
388 54 of the sites are then added into the inversion system. The impact of the CONTRAL data on
389 the inverted carbon fluxes and posterior uncertainties in China and its surrounding areas are
390 quantified. Results show that when the CONTRAIL CO₂ data are added, the inverted carbon
391 sink in China increases, and that in South and Southeast Asia decreases. The changes in South
392 and Southeast Asia make the inverted carbon sinks more comparable with previous studies.
393 CONTRAIL CO₂ data also make a large impact on the inverted inter-annual variation of
394 carbon sinks in China, with the largest change in 2007. This change makes the carbon sink in
395 northern China better correlated with the annual mean air temperature and that in southern
396 China better correlated with the solar radiation and precipitation. Moreover, we use the CO₂
397 data measured at three China observation sites to evaluate the forward simulation results
398 using the TM5 model based on the posterior fluxes. Results show that the large change of the
399 land sink in China in 2007 has made the simulated concentrations in better agreement with the
400 observations at three sites that are not used in the inversion. Finally, it is interesting to note
401 that more CO₂ measurements in or around China added to the inversion lead to an increase in
402 the inverted sinks in China.

403

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412

413 **References**

- 414 Baker, D. F., Law, R. M., Gurney, K. R., Rayner, P., Peylin, P., Denning, A. S., Bousquet, P.,
415 Bruhwiler, L., Chen, Y. H., Ciais, P., Fung, I. Y., Heimann, M., John, J., Maki, T.,
416 Maksyutov, S., Masarie, K., Prather, M., Pak, B., Taguchi, S., and Zhu, Z.: TransCom 3
417 inversion intercomparison: Impact of transport model errors on the interannual variability
418 of regional CO₂ fluxes, 1988–2003, *Global Biogeochem. Cycles*, 20(1), GB1002, 2006.
- 419 Brenninkmeijer, C. A. M., Crutzen, P., Boumard, F., Dauer, T., Dix, B., Ebinghaus, R.,
420 Filippi, D., Fischer, H., Franke, H., Frieß, U., Heintzenberg, J., Helleis, F., Hermann, M.,
421 Kock, H. H., Koepfel, C., Lelieveld, J., Leuenberger, M., Martinsson, B. G., Miemczyk,
422 S., Moret, H. P., Nguyen, H. N., Nyfeler, P., Oram, D., O'Sullivan, D., Penkett, S., Platt,
423 U., Pucek, M., Ramonet, M., Randa, B., Reichelt, M., Rhee, T. S., Rohwer, J., Rosenfeld,
424 K., Scharffe, D., Schlager, H., Schumann, U., Slemr, F., Sprung, D., Stock, P., Thaler, R.,
425 Valentino, F., van Velthoven, P., Waibel, A., Wandel, A., Waschitschek, K.,
426 Wiedensohler, A., Xueref-Remy, I., Zahn, A., Zech, U., and Ziereis, H.: Civil Aircraft for
427 the regular investigation of the atmosphere based on an instrumented container: The new
428 CARIBIC system, *Atmos. Chem. Phys.*, 7, 4953-4976, doi:10.5194/acp-7-4953-2007,
429 2007.
- 430 Boden, T. A., Marland, G., and Andres, R. J.: Global, regional, and national fossil-fuel CO₂
431 emissions, Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory,
432 US Department of Energy, Oak Ridge, Tenn., USA, doi:10.3334/CDIAC/00001_V2010,
433 2010.
- 434 Buitenhuis, E., Le Quéré, C., Aumont, O., Beaugrand, G., Bunker, A., Hirst, A., Ikeda, T.,
435 O'Brien, T., Piontkovski, S., and Straile, D.: Biogeochemical fluxes through
436 mesozooplankton, *Global Biogeochem. Cycles*, 20, GB2003, doi:10.1029/2005GB002511,
437 2006.
- 438 Cao, M. K., Prince, S. D., Li, K. R., Tao, B., Small, J., and Shao, X.M.: Response of
439 terrestrial carbon uptake to climate interannual variability in China, *Global Change*
440 *Biology*, 9, 536–546, 2003.
- 441 CarbonTraker 2010: Fossil Fuel Module,
442 http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2010/documentation_ff.html#ct_doc
443 c, last access: 12 Mar 2014.
- 444 Chen, J. M., Liu, J., Cihlar, J., and Goulden, M. L.: Daily canopy photosynthesis model
445 through temporal and spatial scaling for remote sensing applications, *Ecological*
446 *Modelling*, 124, 99-119, 1999.
- 447 Ciais, P., Reichstein, M., Viovy, N., Granier, A., Ogee, J., Allard, V., Aubinet, M., Buchmann,
448 N., Bernhofer, C., Carrara, A., Chevallier, F., De Noblet, N., Friend, A. D., Friedlingstein,
449 P., Grunwald, T., Heinesch, B., Keronen, P., Knohl, A., Krinner, G., Loustau, D., Manca,
450 G., Matteucci, G., Miglietta, F., Ourcival, J. M., Papale, D., Pilegaard, K., Rambal, S.,
451 Seufert, G., Soussana, J. F., Sanz, M. J., Schulze, E. D., Vesala, T., and Valentini, R.:

452 Europe-wide reduction in primary productivity caused by the heat and drought in 2003,
 453 Nature, 437, 529–533, 2005.

454 Crevoisier, C., Sweeney, C., Gloor, M., Sarmiento, J. L., and Tans, P. P.: Regional US
 455 carbon sinks from three-dimensional atmospheric CO₂ sampling, Proc. Natl. Acad. Sci. U.
 456 S. A., 107(43), 18,348–18,353, doi:10.1073/pnas.0900062107, 2010.

457 Deng, F. and Chen, J. M.: Recent global CO₂ flux inferred from atmospheric CO₂
 458 observations and its regional analyses, Biogeosciences, 8, 3263–3281,
 459 doi:10.5194/bg-8-3263-2011, 2011.

460 Dunn, A. L., Barford, C. C., Wofsy, S. C., Goulden, M. L. and Daube, B. C.: A long-term
 461 record of carbon exchange in a boreal black spruce forest: means, responses to interannual
 462 variability, and decadal trends. Global Change Biology, 13: 577–590. doi:
 463 10.1111/j.1365-2486.2006.01221.x, 2007.

464 Enting, I. G. and Mansbridge, J. V.: Seasonal sources and sinks of atmospheric CO₂: direct
 465 inversion of filtered data, Tellus B, 41, 111–126, 1989.

466 Fang, J. Y., Guo, Z. D., Piao, S. L., and Chen, A. P.: Terrestrial vegetation carbon sinks in
 467 China, 1981–2000, Science in China (D-Earth Science), 50, 1341–1350, 2007.

468 GLOBALVIEW-CO2: Cooperative Atmospheric Data Integration Project – Carbon Dioxide,
 469 NOAA ESRL, Boulder, Colorado, available at:
 470 <http://www.esrl.noaa.gov/gmd/ccgg/globalview/>, last access: 12 Mar 2014, 2010.

471 Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Baker, D., Bousquet, P., Bruhwiler,
 472 L., Chen, Y. H., Ciais, P., Fan, S.M., Fung, I. Y., Gloor, M., Heimann, M., Higuchi, K.,
 473 John, J., Kowalczyk, E., Maki, T., Maksyutov, S., Peylin, P., Prather, M., Pak, B. C.,
 474 Sarmiento, J., Taguchi, S., Takahashi, T., and Yuen, C. W.: TransCom 3 CO₂ inversion
 475 intercomparison: 1. Annual mean control results and sensitivity to transport and prior flux
 476 information. Tellus 55B, 555–579, 2003.

477 Holtslag, A. A.M. and Moeng, C.-H.: Eddy diffusivity and countergradient transport in the
 478 convective atmospheric boundary layer, J. Atmos. Sci., 48, 1690–1698, 1991.

479 Intergovernmental Panel on Climate Change (IPCC), I.S.A.: The Physical Science Basis of
 480 Climate Change: changes in Atmospheric Constituents and in Radiative Forcing.
 481 Cambridge University Press: New York, 2007.

482 Jiang, F., Wang, H.M., Chen, J.M., Zhou, L.X., Ju, W.M., Ding, A.J., Liu, L.X., and Peters,
 483 W.: Nested Atmospheric Inversion for the Terrestrial Carbon Sources and Sinks in China,
 484 Biogeosciences, 10, 5311–5324, doi:10.5194/bg-10-5311-2013, 2013.

485 Ju, W.M., Chen, J.M., Black T.A., Barr A.G., Liu, J., and Chen, B.Z.: Modelling multi-year
 486 coupled carbon and water fluxes in a boreal aspen forest. Agricultural and Forest
 487 Meteorology, 140, 136–151, 2006.

488 Krol, M., Houweling, S., Bregman, B., van den Broek, M., Segers, A., van Velthoven, P.,
 489 Peters, W., Dentener, F., and Bergamaschi, P.: The two-way nested global chemistry -
 490 transport zoom model TM5: algorithm and applications, Atmos. Chem. Phys., 5, 417–432,

- 491 doi:10.5194/acp-5-417-2005, 2005.
- 492 Le Quéré, C., Raupach, M. R., Canadell, J. G., Marland, G., Bopp, L., Ciais, P., Conway, T. J.,
493 Doney, S. C., Feely, R., Foster, P., Friedlingstein, P., Gurney, K., Houghton, R. A., House,
494 J. I., Huntingford, C., Levy, P. E., Lomas, M. R., Majkut, J., Metzl, N., Ometto, J. P.,
495 Peters, G. P., Prentice, I. C., Randerson, J. T., Running, S. W., Sarmiento, J. L., Schuster,
496 U., Sitch, S., Takahashi, T., Viovy, N., van der Werf, G. R., and Woodward, F. I.: Trends
497 in the sources and sinks of carbon dioxide, *Nat. Geosci.*, 2, 831–836,
498 doi:10.1038/ngeo689, 2009.
- 499 Liu, L. X., Zhou, L. X., Zhang, X. C., Wen, M., Zhang, F., Yao, B., and Fang, S. X.: The
500 characteristics of atmospheric CO₂ concentration variation of four national background
501 stations in China, *Sci. China Ser. D-Earth Sci.*, 52, 1857–1863, doi:
502 10.1007/s11430-009-0143-7, 2009.
- 503 Louis, J. F.: A parametric model of vertical eddy fluxes in the atmosphere, *Boundary Layer*
504 *Meteor.*, 17, 187–202, 1979.
- 505 Machida, T., Matsueda, H., Sawa, Y., Nakagawa, Y., Hirokuni, K., Kondo, N., Goto, K.,
506 Nakazawa, T., Ishikawa, K., and Ogawa T.: Worldwide measurements of atmospheric
507 CO₂ and other trace gas species using commercial airlines, *J. Atmos. Oceanic Technol.*,
508 25, 1744–1754, doi:10.1175/2008JTECHA1082.1, 2008.
- 509 Machida, T., Tohjima, Y., Katsumata, K., and Mukai, H.: A new CO₂ calibration scale based
510 on gravimetric one-step dilution cylinders in National Institute for Environmental Studies
511 -NIES 09 CO₂ scale, GAW Rep. 194, pp. 114–119, World Meteorol. Organ., Geneva,
512 Switzerland, 2011.
- 513 Masarie, K. A., and Tans, P. P.: Extension and integration of atmospheric carbon dioxide data
514 into a globally consistent measurement record, *J. Geophys. Res.*, 100(D6), 11,593–11, 610,
515 doi:10.1029/95JD00859, 1995.
- 516 Matsueda, H., Machida, T., Sawa, Y., Nakagawa, Y., Hirokuni, K., Ikeda, H., Kondo, N., and
517 Goto, K.: Evaluation of atmospheric CO₂ measurements from new flask air sampling of
518 JAL airliner observations, *Pap. Meteorol. Geophys.*, 59, 1–17, doi:10.2467/mripapers.
519 59.1, 2008.
- 520 Myoung, B., Choi, Y. S., Hong, S., and Park, S. K.: Inter- and intra-annual variability of
521 vegetation in the Northern Hemisphere and its association with precursory meteorological
522 factors, *Global Biogeochem. Cycles*, 27, 31–42, doi:10.1002/gbc.20017, 2013.
- 523 Niwa, Y., Machida, T., Sawa, Y., Matsueda, H., Schuck, T. J., Brenninkmeijer, C. A. M.,
524 Imasu, R., and Satoh, M.: Imposing strong constraints on tropical terrestrial CO₂ fluxes
525 using passenger aircraft based measurements, *J. Geophys. Res.*, 117, D11303,
526 doi:10.1029/2012 JD017474, 2012.
- 527 Olivier, J.G.J. and Berdowski J.J.M.: Global emissions sources and sinks. In: Berdowski, J.,
528 Guicherit, R. and B.J. Heij (eds.) *The Climate System*, pp. 33-78. A.A. Balkema
529 Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands. ISBN 90 5809 255 0,

530 2001.

531 Pacala, S. W., Hurtt, G. C., Baker, D., Peylin, P., Houghton, R. A., Birdsey, R. A., Heath, L.,
532 Sundquist, E. T., Stallard, R. F., Ciais, P., Moorcroft, P., Caspersen, J. P., Shevliakova, E.,
533 Moore, B., Kohlmaier, G., Holland, E., Gloor, M., Harmon, M. E., Fan, S.-M., Sarmiento,
534 J. L., Goodale, C. L., Schimel, D., and Field, C. B.: Consistent land- and atmosphere-based
535 US carbon sink estimates. *Science*, 292, 2316-2320, 2001.

536 Pan, Y., Birdsey, R. A., Fang, J., Houghton, R., Kauppi, P. E., Kurz, W. A., Phillips, O. L.,
537 Shvidenko, A., Lewis, S. L., Canadell, J. G., Ciais, P., Jackson, R. B., Pacala, S., McGuire,
538 A. D., Piao, S., Rautiainen, A., Sitch, S., and Hayes, D.: A large and persistent carbon
539 sink in the world's forests, *Science*, 333, 988–993, doi:10.1126/science.1201609, 2011.

540 Patra, P. K., Niwa, T. J. Schuck, C. A. M. Brenninkmeijer, T. Machida, H. Matsueda, and
541 Y. Sawa: Carbon balance of South Asia constrained by passenger aircraft CO₂
542 measurements, *Atmos. Chem. Phys.*, 11, 4163-4175, 2011.

543 Patra, P. K., Canadell, J. G., Houghton, R. A., Piao, S. L., Oh, N.-H., Ciais, P., Manjunath, K.
544 R., Chhabra, A., Wang, T., Bhattacharya, T., Bousquet, P., Hartman, J., Ito, A., Mayorga,
545 E., Niwa, Y., Raymond, P. A., Sarma, V. V. S. S., and Lasco, R.: The carbon budget of
546 South Asia, *Biogeosciences*, 10, 513-527, doi:10.5194/bg-10-513-2013, 2013.

547 Piao, S. L., Fang, J. Y., Ciais, P., Peylin, P., Huang, Y., Sitch, S., and Wang, T.: The carbon
548 balance of terrestrial ecosystems in China, *Nature*, 458, 1009–1013,
549 doi:10.1038/nature07944, 2009.

550 Rayner, N. A., Parker, D. E., Horton, E. B., Folland, C. K., Alexander, L. V., Rowell, D. P.,
551 Kent, E. C., Kaplan, A.: Global analyses of sea surface temperature, sea ice, and night
552 marine air temperature since the late nineteenth century, *J. Geophys. Res.*, 108 (D14), doi:
553 10.1029/2002JD002670, 2003.

554 Rayner, P. J., Enting, I. G., Francey, R. J. and Langenfelds, R.: Reconstructing the recent
555 carbon cycle from atmospheric CO₂, ¹³C and O₂/N₂ observations, *Tellus* 51B, 213–232,
556 1999.

557 Russel, G. and Lerner, J.: A new finite-differencing scheme for the tracer transport equation, *J.*
558 *Appl. Meteorol.*, 20, 1483–1498, 1981.

559 Sawa, Y., Machida, T., and Matsueda, H.: Seasonal variations of CO₂ near the tropopause
560 observed by commercial aircraft, *J. Geophys. Res.*, 113, D23301,
561 doi:10.1029/2008JD010568, 2008.

562 Schuck, T. J., Brenninkmeijer, C. A. M., Slemr, F., Xueref-Remy, I., and Zahn, A.:
563 Greenhouse gas analysis of air samples collected onboard the CARIBIC passenger aircraft,
564 *Atmos. Meas. Tech.*, 2, 449–464, doi:10.5194/amt-2-449-2009, 2009.

565 Stephens, B. B., Gurney, K. R., Tans, P. P., Sweeney, C., Peters, W., Bruhwiler, L., Ciais, P.,
566 Ramonet, M., Bousquet, P., Nakazawa, T., Aoki, S., Machida, T., Inoue, G., Vinnichenko,
567 N., Lloyd, J., Jordan, A., Heimann, M., Shibistova, O., Langenfelds, R. L., Steele, L. P.,
568 Francey, R. J., and Denning, A. S.: Weak northern and strong tropical land carbon uptake

569 from vertical profiles of atmospheric CO₂, *Science*, 316, 1732–1735, 10.1126/science.
570 1137004, 2007.

571 Tian, H., Melillo, J., Lu, C.Q., Kicklighter, D., Liu, M.L., Ren, W., Xu, X.F., Chen, G.S.,
572 Zhang, C., Pan, S.F., Liu, J.Y., and Running, S.: China's terrestrial carbon balance:
573 Contributions from multiple global change factors, *Global Biogeochem. Cycles*, 25,
574 GB1007, doi:10.1029/2010GB003838, 2011.

575 Tiedtke, M.: A comprehensive mass flux scheme for cumulus parameterisation in large scale
576 models, *Mon. Wea. Rev.*, 177, 1779–1800, 1989.

577 van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S.,
578 Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and
579 the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009),
580 *Atmos. Chem. Phys.*, 10, 11707–11735, doi:10.5194/acp-10-11707–2010, 2010.

581 Xueref-Remy, I., Messager, C., Filippi, D., Pastel, M., Nedelec, P., Ramonet, M., Paris, J. D.,
582 and Ciais, P.: Variability and budget of CO₂ in Europe: Analysis of the CAATER airborne
583 campaigns—Part 1: Observed variability, *Atmos. Chem. Phys.*, 11, 5655–5672,
584 doi:10.5194/acp-11-5655-2011, 2011.

585 Yan, J. H., Zhang, Y. P., Yu, G. R., Zhou, G. Y., Zhang, L. M., Li, K., Tan, Z. H., and Sha, L.
586 Q.: Seasonal and inter-annual variations in net ecosystem exchange of two old-growth
587 forests in southern China. *Agricultural and Forest Meteorology*, 182-183, 257-265, Doi:
588 10.1016/j.agrformet.2013.03.002, 2013.

589 Yuan, W., Luo, Y., Richardson, A. D., Oren, R., Luysaert, S., Janssens, I. A., Ceulemans, R.,
590 Zhou, X., Grünwald, T., Aubinet, M., Berhofer, C., Baldocchi, D. D., Chen, J., Dunn, A.
591 L., Deforest, J. L., Dragoni, D., Goldstein, A. H., Moors, E., William Munger, J., Monson,
592 R. K., Suyker, A. E., Starr, G., Scott, R. L., Tenhunen, J., Verma, S. B., Vesala, T. and
593 Wofsy, S. C.: Latitudinal patterns of magnitude and interannual variability in net
594 ecosystem exchange regulated by biological and environmental variables. *Global Change*
595 *Biology*, 15: 2905–2920, doi: 10.1111/j.1365-2486.2009.01870.x, 2009.

596 Zhu, W.Q., Pan, Y.Z., Yang, X.Q., and Song, G.B.: Comprehensive analysis of the impact of
597 climatic changes on Chinese terrestrial net primary productivity, *Chinese Science Bulletin*,
598 52(23), 3253-3260, 2007.

599 Zou, X.K., Ren, G.Y., and Zhang, Q.: Droughts Variations in China Based on a Compound
600 Index of Meteorological Drought, *Climatic and Environmental Research*, 15 (4), 371–378,
601 2010. (in Chinese)

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604 **List of Figure Captions**

605 **Figure 1.** A map of CONTRAIL measurements; (a) the CONTRAIL measurement locations
606 (red dots), black rectangles represent the area partitions of the measurements of level flights,
607 shaded represent different inversion regions over Eurasia; (b) Time series of the
608 measurements over East China Sea, black line indicates a curve fitted to the daily CO₂.

609 **Figure 2.** An inversion scheme: 21 regions in Asia (13 regions in China) and 22 regions for
610 the rest of the globe. Locations of 184 CO₂ observational sites are also indicated, including
611 130 sites from GV dataset (54 flask sites, 7 continuous sites, 5 tower sites, 6 ship sites, 58
612 aircraft sites) and 54 sites from CONTRAIL aircraft measurements (bold ones include 3
613 vertical sites at 2000–4000, 4000–6000, 6000–8000 m for ascending and descending flights
614 data and thin ones include 1 sites at 8000-12000 m for level flights data).

615 **Figure 3.** Locations of observations (black point: meteorological data locations; red triangle:
616 CO₂ observation sites in China, which are used for evaluation in this study)

617 **Figure 4.** Impact on inter-annual variations of inverted carbon flux and posterior uncertainty
618 in China

619 **Figure 5.** Inter-annual variations of the posterior fluxes and climate factors in (a) southern
620 China and (b) northern China

621 **Figure 6.** Inverted global carbon flux for a) Case GV, unit: gC m⁻²yr⁻¹; b) Case GVCT, unit:
622 gC m⁻²yr⁻¹; and impact of CONTRAIL CO₂ on c) the inverted carbon fluxes, Case GVCT –
623 Case GV, unit: gC m⁻²yr⁻¹, and d) posterior uncertainties, (Case GV-Case GVCT)×100/Case
624 GV, unit: %; averaged for 2006 to 2008.

625 **Figure 7.** Mean monthly fluxes in a) China, b) South Asia and c) Southeast Asia during 2006
626 – 2008 (Case GV: only constrained by GV CO₂; Case GVCT: constrained by both GV CO₂
627 and CONTRAIL CO₂)

628 **Figure 8.** Contributions from emissions of different regions in July 2007 and the previous
629 five months (1 PgC month⁻¹ region⁻¹) to the CO₂ concentration in July 2007 at 2000 – 4000 m
630 over Taipei airport (TPE)

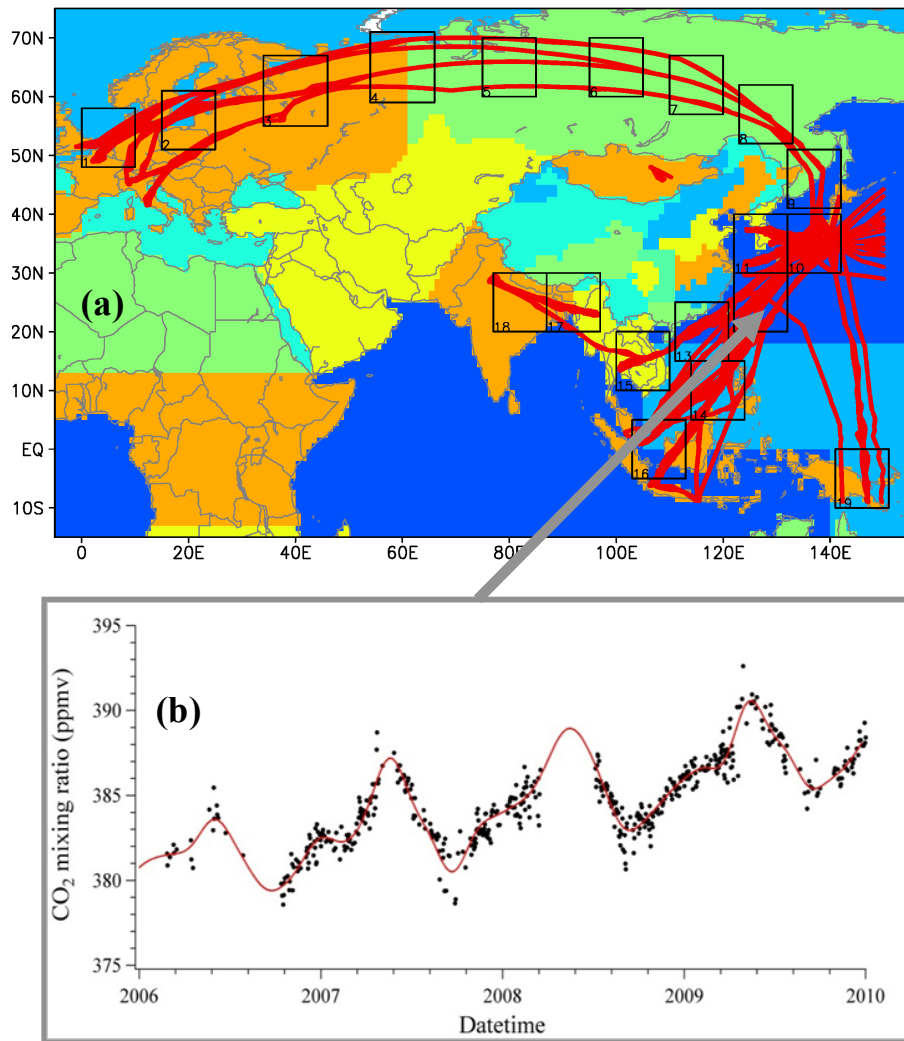
631 **Figure 9.** Simulated and observed monthly CO₂ concentrations at three China stations, the
632 black lines represent the observations, the orange lines represent the simulations from the land

633 fluxes constrained with GV CO₂ only, the blue lines represent the simulations from the land
634 fluxes constrained by additional CONTRAIL CO₂, and the numbers represent the
635 concentration amplitude between wintertime and summertime

636 **Figure 10.** The sensitivity of the influence of (a) monthly prior flux uncertainty on the
637 inverted carbon sinks over China, including (b) the inter-annual variations and (c) the
638 monthly variations, as well as on (d) the simulated CO₂ concentrations in 2007 at the three
639 Chinese sites using the inverted carbon fluxes.

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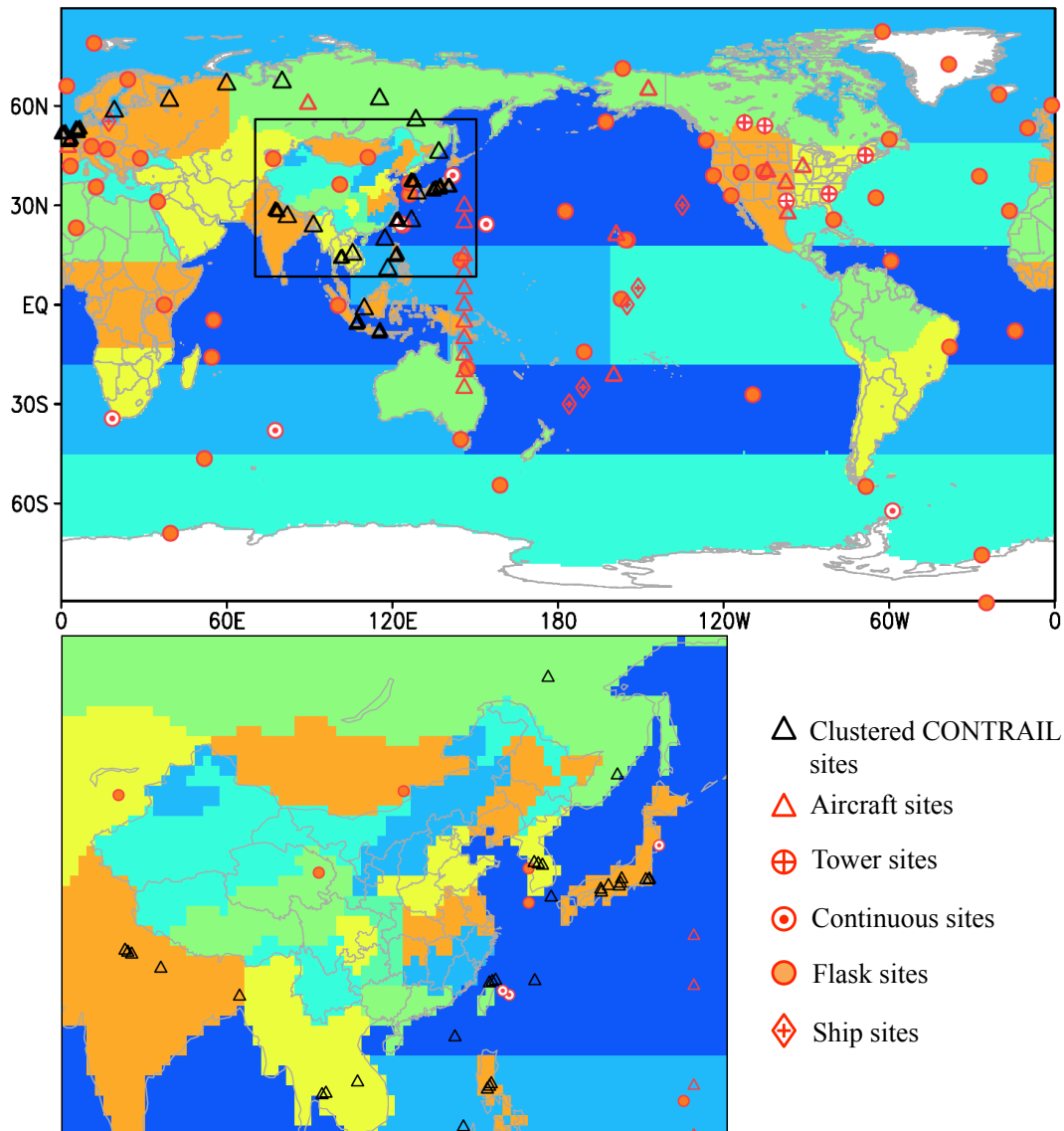
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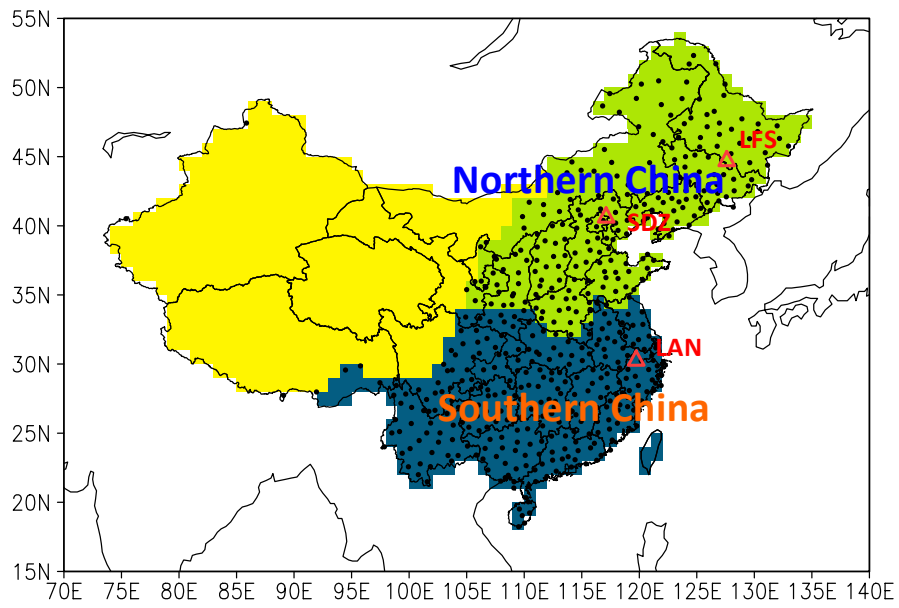


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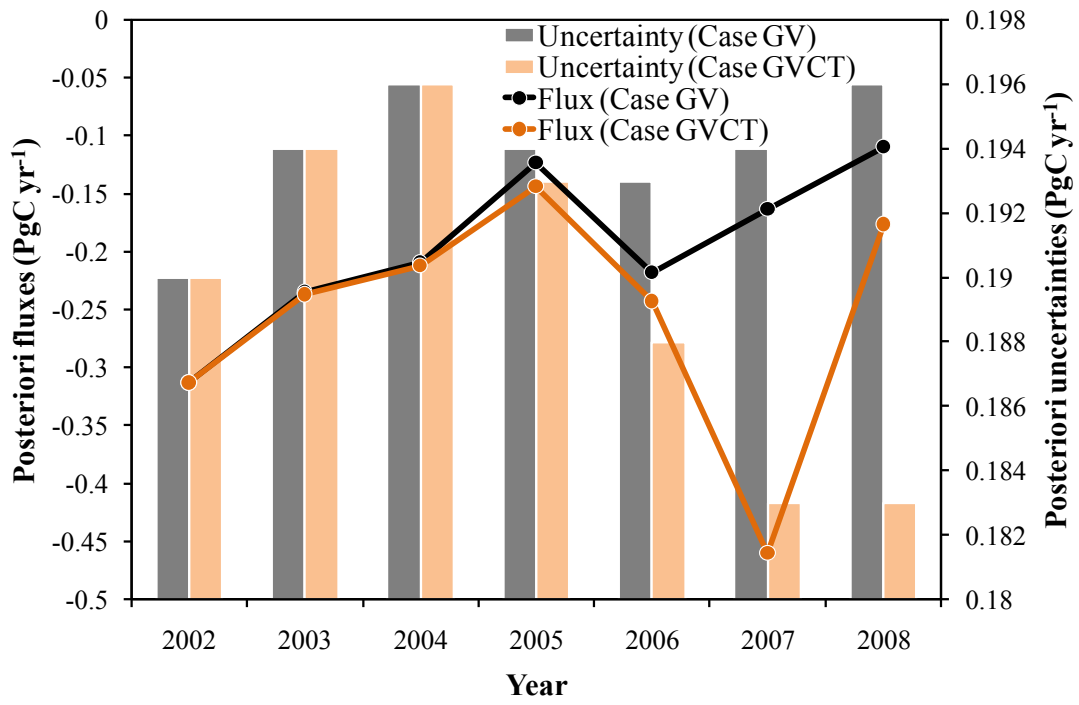


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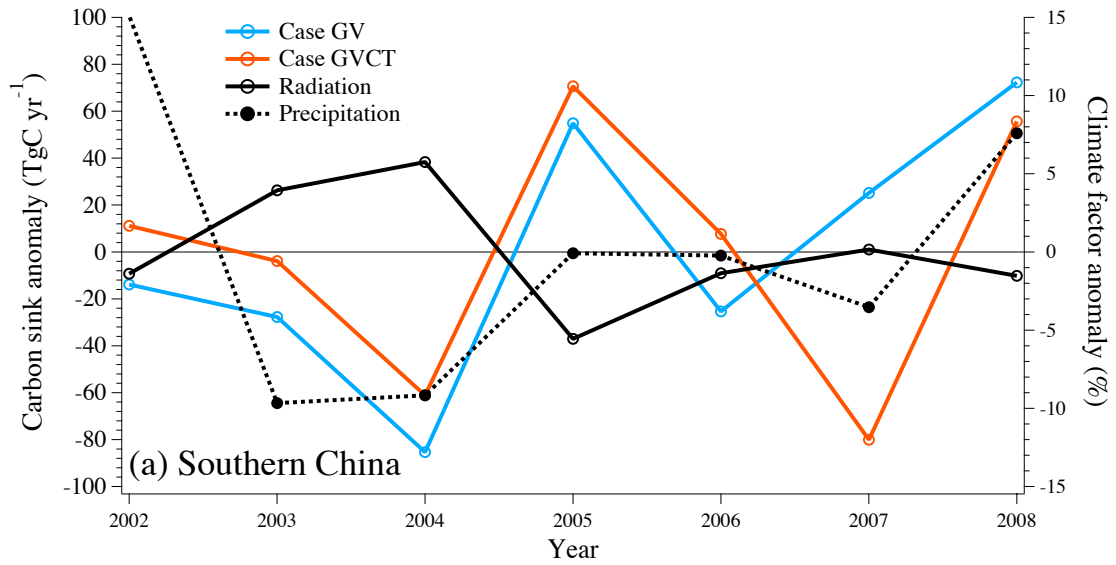
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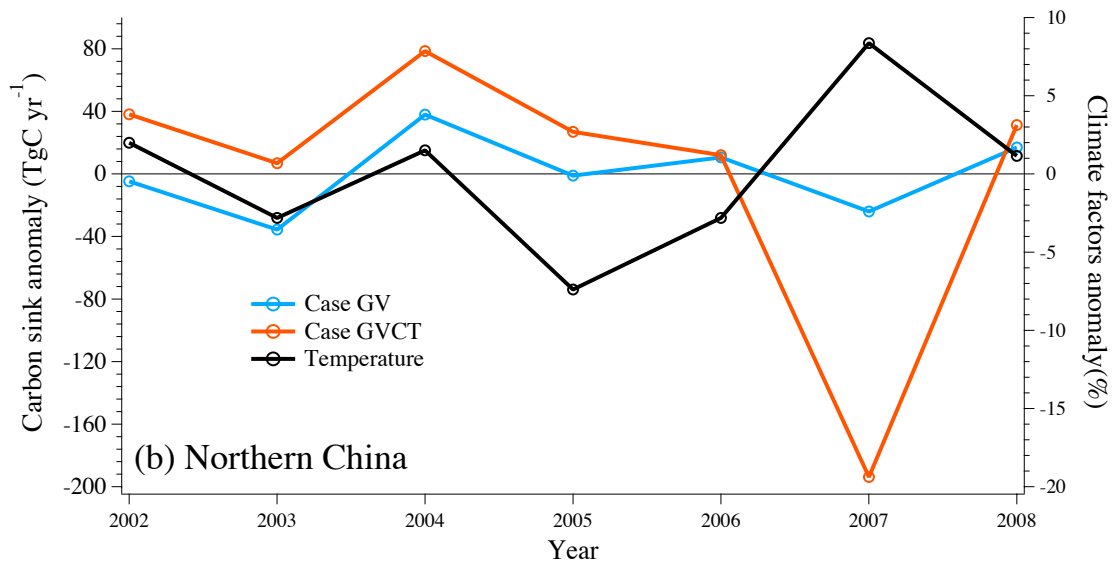
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675 **Figure 5.** Inter-annual variations of the posterior fluxes and climate factors in (a) southern

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China and (b) northern China

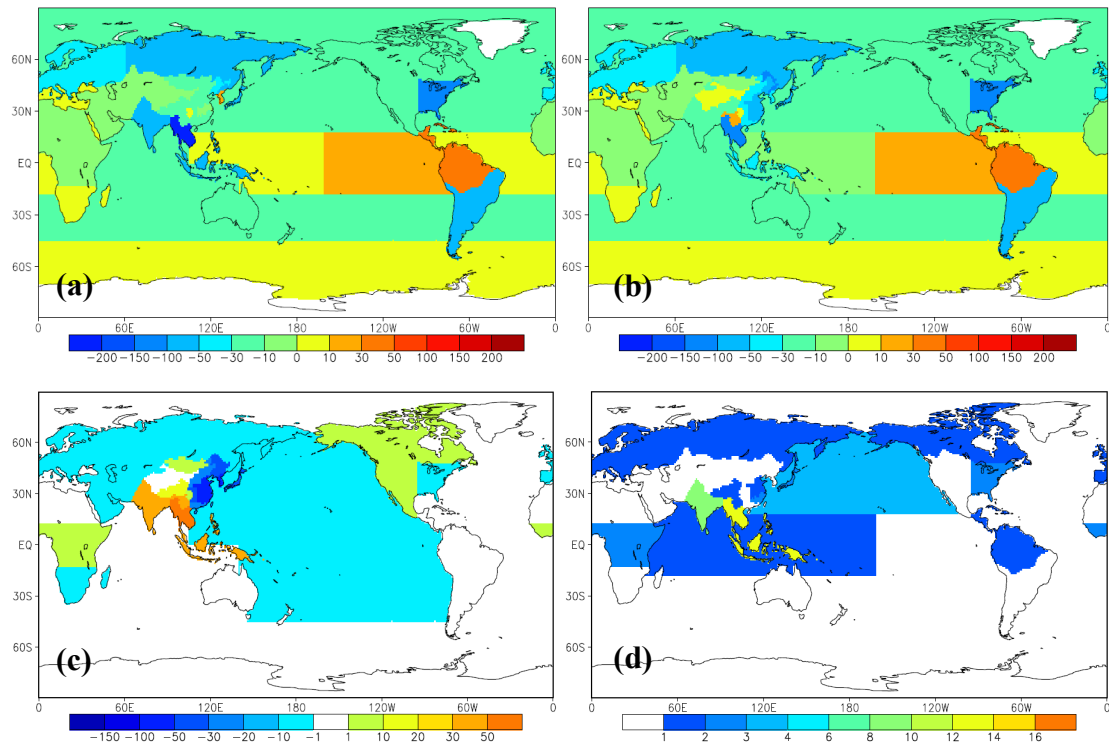
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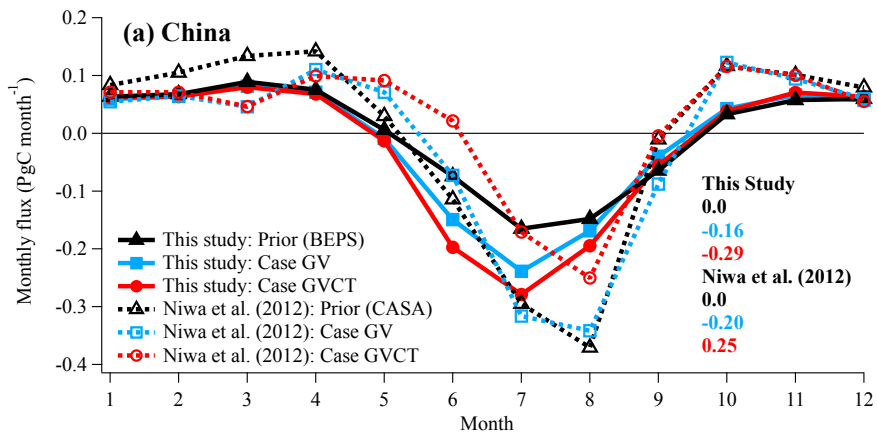
683

684 **Figure 6.** Inverted global carbon flux for a) Case GV, unit: $\text{gC m}^{-2} \text{yr}^{-1}$; b) Case GVCT, unit:685 $\text{gC m}^{-2} \text{yr}^{-1}$; and impact of CONTRAIL CO_2 on c) the inverted carbon fluxes, Case GVCT -686 Case GV, unit: $\text{gC m}^{-2} \text{yr}^{-1}$ and d) posterior uncertainties, $(\text{Case GV} - \text{Case GVCT}) \times 100 / \text{Case}$

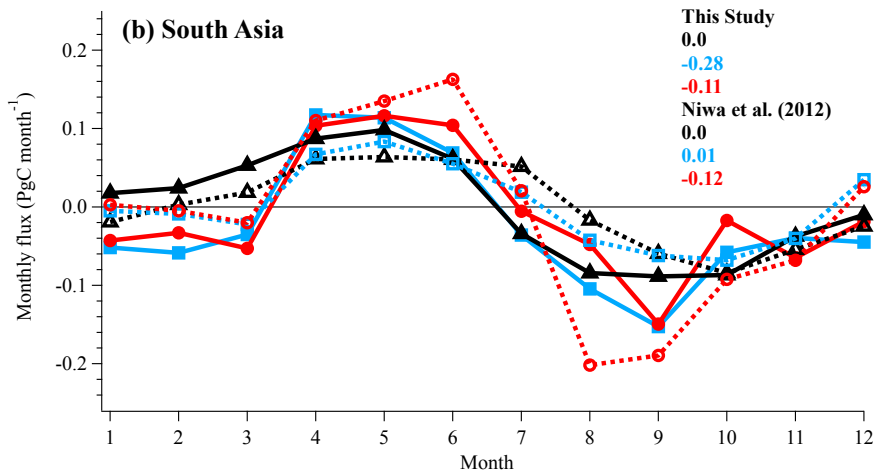
687 GV, unit: %; averaged for 2006 to 2008.

688

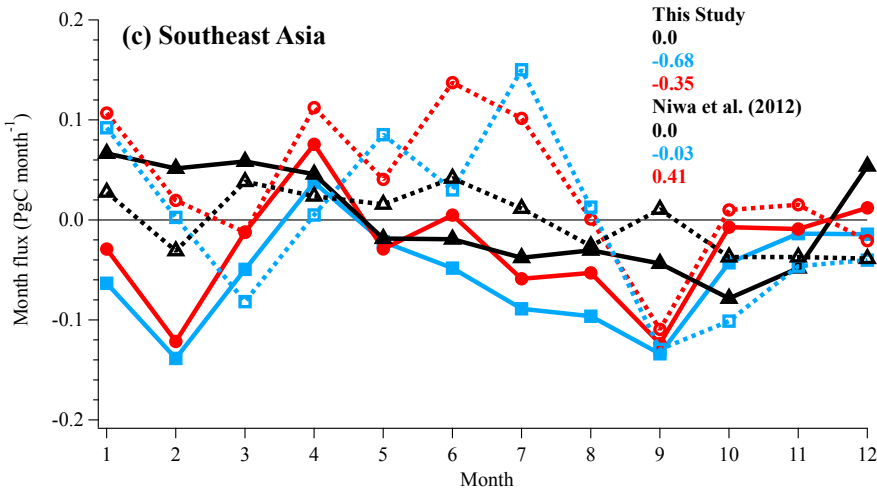
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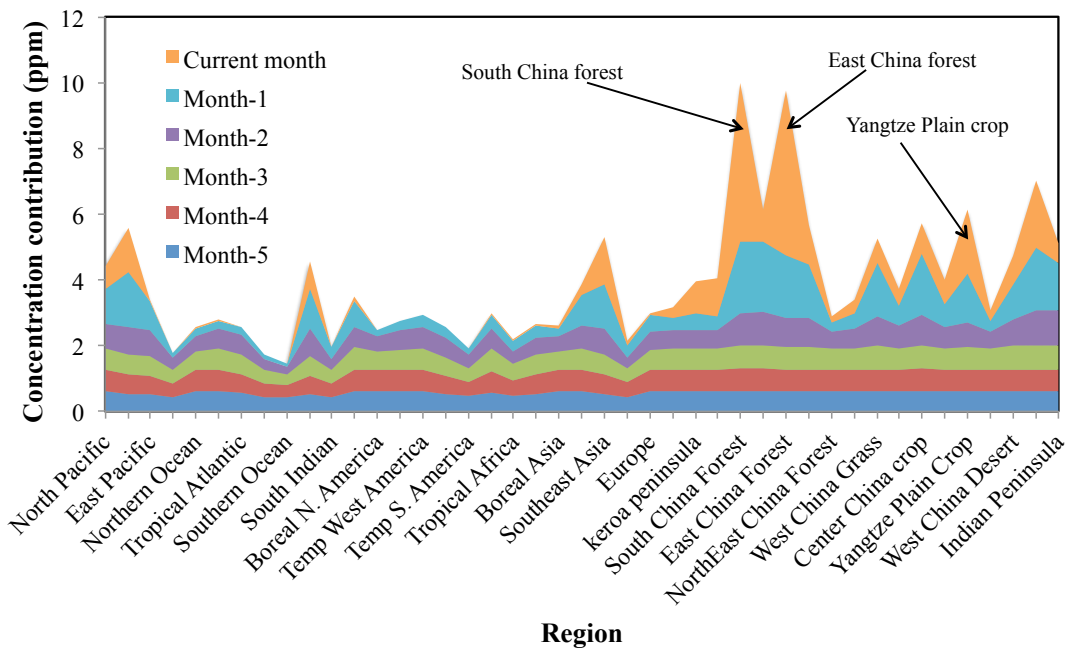
693 **Figure 7.** Mean monthly fluxes in a) China, b) South Asia and c) Southeast Asia during 2006

694 – 2008 (Case GV: only constrained by GV CO₂; Case GVCT: constrained by both GV CO₂

695 and CONTRAIL CO₂)

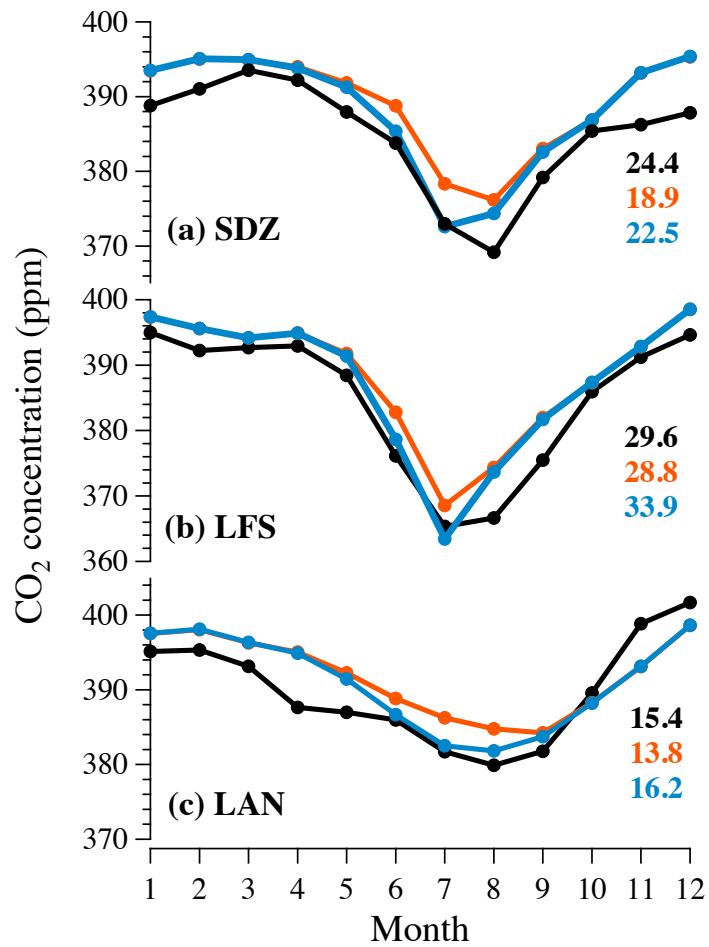
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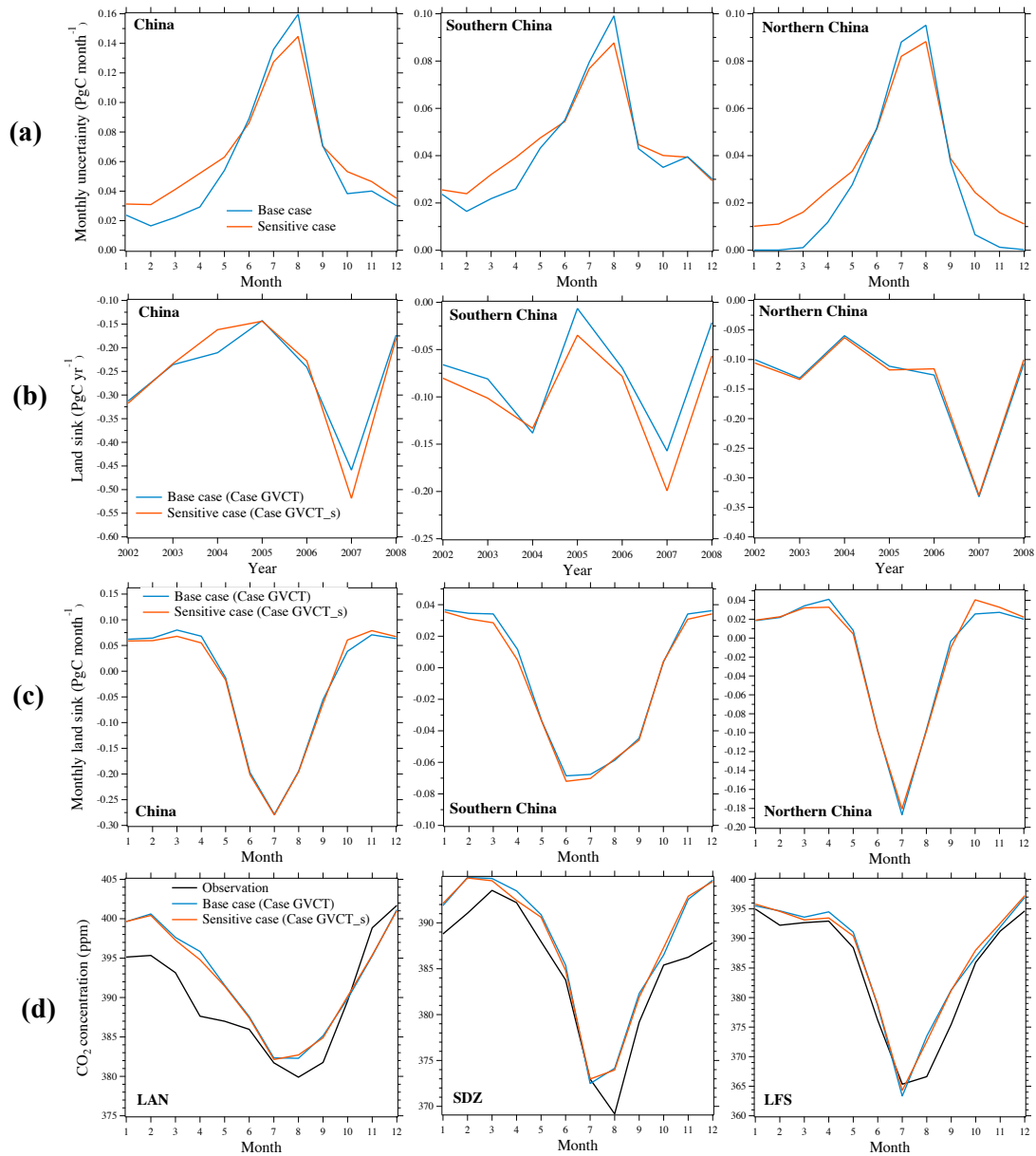
698

699 **Figure 8.** Contributions from emissions of different regions in July 2007 and the previous
700 five months ($1 \text{ PgC month}^{-1} \text{ region}^{-1}$) to the CO₂ concentration in July 2007 at 2000 – 4000 m
701 over Taipei airport (TPE)



703

704 **Figure 9.** Simulated and observed monthly CO₂ concentrations at three China stations in
 705 2007, the black lines represent the observations, the orange lines represent the simulations
 706 from the land fluxes constrained with GV CO₂ only, the blue lines represent the simulations
 707 from the land fluxes constrained by additional CONTRAIL CO₂, and the numbers represent
 708 the concentration amplitude between wintertime and summertime



710

711 **Figure 10.** The sensitivity of the influence of (a) monthly prior flux uncertainty on the
 712 inverted carbon sinks over China, including (b) the inter-annual variations and (c) the
 713 monthly variations, as well as on (d) the simulated CO₂ concentrations in 2007 at the three
 714 Chinese sites using the inverted carbon fluxes.