

# Gradients of Column CO<sub>2</sub> across North America from the NOAA Global Greenhouse Gas Reference Network

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**Abstract.** This study analyzes seasonal and spatial patterns of column carbon dioxide (CO<sub>2</sub>) over North America calculated from aircraft and tall tower measurements from the NOAA Global Greenhouse Gas Reference Network from 2004 to 2014. Consistent with expectations, gradients between the eight regions studied are larger below 2 km than above 5 km. The 11-year mean CO<sub>2</sub> dry mole fraction (XCO<sub>2</sub>) in the column below ~330 hPa (~ 8 km above sea level) from NOAA's CO<sub>2</sub> data assimilation model, CarbonTracker (CT2015), demonstrates good agreement with those calculated from calibrated measurements on aircraft and towers. Total column XCO<sub>2</sub> was attained by combining modeled CO<sub>2</sub> above 330 hPa from CT2015 with the measurements. We find large spatial gradients of total column XCO<sub>2</sub> during June to August, with north and northeast regions having ~3 ppm stronger summer drawdown (peak to valley amplitude in seasonal cycle) than the south and southwest regions. The long-term averaged spatial gradients of total column XCO<sub>2</sub> across North America show a smooth pattern that mainly reflects the large-scale circulation. We have conducted a CarbonTracker experiment to investigate the impact of Eurasian long-range transport. The result suggests that the large summer time Eurasian boreal flux contributes about half of the north-south column XCO<sub>2</sub> gradient across North America. Our results confirm that continental-scale total column XCO<sub>2</sub> gradients simulated by CarbonTracker are realistic and can be used to evaluate the credibility of some spatial patterns from satellite retrievals, such as the long term average of growing-season spatial patterns from satellite retrievals reported for Europe which show larger spatial difference (~ 6 ppm) and scattered hot spots.

## 1 Introduction

Atmospheric measurements of carbon dioxide (CO<sub>2</sub>) from ground and airborne platforms have greatly increased our knowledge of the global carbon cycle. Observations of CO<sub>2</sub>, including the NOAA Global Greenhouse Gas Reference Network (GGGRN), initially emphasized ground-based measurements. These observations, started by C.D. Keeling, have monitored the CO<sub>2</sub> trend on both regional and global scales for over 50 years (e.g., Keeling and Rakestraw, 1960; Tans et al., 1989). In addition, the frequency and spatial distribution of airborne measurements have increased rapidly in the last two decades, providing important information about horizontal and vertical variability of atmospheric CO<sub>2</sub> (e.g., Gerbig et al., 2003; Choi et al., 2008; Biraud et al., 2013). Routine aircraft

36 measurements from the NOAA/ESRL GGGRN monitor the large-scale distributions of a suite of trace gases,  
37 including CO<sub>2</sub>, under the influence of continental processes (Sweeney et al., 2015). A very successful approach has  
38 been to employ commercial aircraft as a platform for CO<sub>2</sub> measurements, such as Japan's CONTRAIL  
39 (Comprehensive Observation Network for TRace gases by AIRliner) project, which has provided valuable  
40 information for CO<sub>2</sub> in the high troposphere and lower stratosphere (Machida et al., 2002; Machida et al., 2008).  
41 Vertical profiles of atmospheric CO<sub>2</sub> reflect the combined influences of surface fluxes and atmospheric mixing.  
42 Vertical profiles are particularly useful for evaluating vertical mixing in atmospheric transport models that are used  
43 for inverse modeling (e.g. Stephens et al., 2007) to derive estimates of regional- to continental-scale CO<sub>2</sub> sources  
44 and sinks (e.g., Tans et al., 1990; Gurney et al., 2002; Gurney et al., 2004; Ciais et al., 2010;).

45 While CO<sub>2</sub> sources and sinks are well constrained at the global scale by global mass balance, it remains  
46 challenging to accurately resolve CO<sub>2</sub> sources and sinks at regional to continental-scale, the apportionment of which  
47 depends on relatively minor variations of the observed spatial and temporal patterns of CO<sub>2</sub>. When averaging over a  
48 few months and longer the largest portion of the variations over continents results from hemispheric-scale terrestrial  
49 uptake/emissions (photosynthesis)/respiration) and fossil fuel emissions, while regional net fluxes can make a  
50 relatively small contribution to the signal. For example, a simple mass balance argument shows that all U.S. CO<sub>2</sub>  
51 emissions from fossil fuel burning (~1.4 Pg yr<sup>-1</sup>) create a total column enhancement of only 0.6 ppm on average in  
52 air parcels over the East Coast compared to the West Coast and Gulf Coast if we assume an average of 5 days for the  
53 winds to flush the contiguous U.S. (~8×10<sup>12</sup> m<sup>2</sup>).

54 With careful calibration, air handling, and analysis, the uncertainties of in-situ measurements are less than 0.1  
55 ppm. However, in-situ observation networks are sparse in global and regional coverage. Remote sensing data  
56 radically increase the number of observations and capture under-sampled regions. It could have a valuable impact on  
57 our understanding of the carbon cycle. However, both the precision and the potential of even very small systematic  
58 biases in remote sensing measurements need to be carefully evaluated, especially those that depend on regional and  
59 seasonal conditions. Vertical profiles from in-situ CO<sub>2</sub> measurements have been used to evaluate ground-based total  
60 column XCO<sub>2</sub> (the "X" stands for dry mole fraction) determinations, such as those from the Total Carbon Column  
61 Observing Network (TCCON) (Washenfelder et al., 2006; Wunch et al., 2010; Messerschmidt et al., 2011; Tanaka  
62 et al., 2012). The uncertainty of TCCON total column CO<sub>2</sub> is reported to be 0.4 ppm (1σ) after comparison to  
63 aircraft measurements (Wunch et al., 2010). Vertical profiles are also used to evaluate other satellite retrievals of  
64 total column XCO<sub>2</sub>, such as those from the Tropospheric Emission Spectrometer (TES)(Kulawik et al., 2013) and  
65 the Greenhouse Gases Observing SATellite (GOSAT) (Inoue et al., 2013, 2016; Saitoh et al., 2016). Satellite  
66 retrieval products have known and unknown biases (due to errors in spectroscopy, viewing geometry, spatial  
67 differences in clouds and aerosols, surface albedo, etc.) that can result in false horizontal gradients in total column  
68 XCO<sub>2</sub> for inverse estimates of sources (Miller et al., 2007; Crisp et al., 2012; Feng et al., 2016). After correction for  
69 known biases, the mean GOSAT total column CO<sub>2</sub> (NIES retrievals) biases range between -2.09 to 3.37 ppm (mean  
70 = 0.11 ppm, S.D.= 1.11 ppm; 20 out of 27 stations show biases lower than 1 ppm) across different aircraft sites over  
71 land when compared with aircraft-based total column XCO<sub>2</sub> (Inoue et al., 2016). The Orbiting Carbon Observatory-  
72 2 (OCO-2) retrieval of total column XCO<sub>2</sub> was estimated to have a mean difference less than 0.5 ppm from TCCON,

73 with RMS differences typically below 1.5 ppm after bias correction (Wunch et al., 2016). The overall uncertainty of  
74 satellite retrievals is relatively large compared with the total column XCO<sub>2</sub> calculated from in-situ measurements.  
75 Total column XCO<sub>2</sub> calculated from vertical profiles from the Japanese CONTRAIL project (Machida et al., 2008)  
76 and from the NOAA Carbon Cycle and Greenhouse Gas aircraft program (Sweeney et al., 2015) complemented with  
77 simulated profiles from a chemistry–transport model above the maximum altitude of the data have uncertainty less  
78 than 1 ppm (Miyamoto et al., 2013). The smaller uncertainty of the in situ-based total column XCO<sub>2</sub> suggests that  
79 they can be used to evaluate satellite retrievals of column averaged CO<sub>2</sub>. Since aircraft profiles co-located with  
80 satellite retrievals are rare, it is useful to consider the statistics of total column XCO<sub>2</sub> fields derived from repeated  
81 aircraft profiles over particular locations.

82 The effect of satellite column averaging kernels and a priori profiles when comparing aircraft-based column  
83 XCO<sub>2</sub> with GOSAT retrievals has been assessed by Inoue et al. (2013). For the case considered, application of the  
84 averaging kernel and a priori profile to simulate total column XCO<sub>2</sub> was generally within  $\pm 0.1$  ppm of the density  
85 weighted total column, suggesting that the averaging kernels can only account for small part of the overall  
86 uncertainty of the GOSAT total column XCO<sub>2</sub> (Inoue et al., 2013).

87 Transparent and objective estimates of CO<sub>2</sub> sources and sinks derived from atmospheric measurements are  
88 essential for validating emissions reduction efforts and other mitigation policies, and for lowering the uncertainties  
89 of carbon cycle-climate feedbacks. The latter are major ambiguities in predicting future climate, such as potential  
90 uncontrolled CH<sub>4</sub> and CO<sub>2</sub> emissions from warming permafrost in Arctic regions. Satellite retrievals of total column  
91 XCO<sub>2</sub> can significantly improve estimates of sources and sinks only if they are sufficiently precise and accurate (   
92 Houweling et al., 2004; Chevallier et al., 2014), meaning that even very small systematic errors (biases) must be  
93 eliminated. Here, we analyze the spatial and temporal variability of column CO<sub>2</sub> over North America using well-  
94 calibrated CO<sub>2</sub> measurements from aircraft and tall tower, and we use model results from NOAA’s CarbonTracker,  
95 version CT2015 (Peters et al. 2007, with updates documented at <http://carbontracker.noaa.gov>) to investigate the  
96 primary drivers of variability in total column XCO<sub>2</sub>. The aircraft data enable direct analysis of column CO<sub>2</sub>  
97 characteristics, which is the fundamental step for accurate apportionment of sources and sinks. This study focuses on  
98 long-term averaged column CO<sub>2</sub> gradients and the contributions of different vertical layers to the total column  
99 variability. It can serve as a reference for evaluating regional and seasonal biases of current and future column CO<sub>2</sub>  
100 retrievals from both ground and satellite platforms.

## 101 **2 Methods**

### 102 **2.1 Aircraft and tall tower sampling**

103 Aircraft sampling in the NOAA GGGRN intends to provide vertical profiles of long-lived trace gases to capture  
104 their seasonal and interannual variability. The aircraft sampling system consists of 12 borosilicate glass flasks in  
105 each programmable flask package (PFP), a stainless-steel gas manifold system, and a data logging and control.  
106 These flasks (0.7 L each) are pressurized to obtain 2.2 L of sample air from each target altitude. Air samples are then  
107 shipped back to NOAA/ESRL for carefully calibrated and quality-controlled measurements. Carbon dioxide is

108 measured using a nondispersive infrared analyzer. Long-term measurements at ~15 sites are carried out using light  
109 aircraft that can reach 8.5 km. Air samples are collected mostly during late morning to early afternoon, when the air  
110 mass within the planetary boundary layer (PBL) is generally well mixed, and CO<sub>2</sub> enhancement near the ground  
111 from plant respiration during the night has been mixed throughout the boundary layer. Normally, the aircraft follows  
112 a pre-decided route such that most samples are collected within 0.1° of the site location. The sampling frequency  
113 varies from site to site, currently from twice a month to once every 1.5 months. For more sampling details, quality  
114 control discussions, and an evaluation of the sampling frequency, please refer to Sweeney et al. (2015). More  
115 information on the aircraft sites can be found at <http://www.esrl.noaa.gov/gmd/ccgg/aircraft/>. We estimate the  
116 uncertainty of individual measurements of CO<sub>2</sub> in flask air (68% confidence level) at 0.08 ppm. However, we have  
117 seen evidence of positive biases for samples collected using older flasks that may contain contaminants. Andrews  
118 et al. (2014) reported biases that increased from <0.1 ppm in 2008 to an average offset in 2013 of 0.36 ppm. The  
119 aircraft sampling protocol was modified starting in August 2014 to mitigate this bias. For samples collected prior the  
120 protocol change, laboratory tests showed that new/clean flasks have zero bias, but some older/dirty flasks could have  
121 biases of > 1 ppm. This bias is not consistent among individual flasks and increasing over time (Andrews et al.,  
122 2014), the potential bias is hard to quantify for measurements before August 2014. Thus, the high bias is not  
123 corrected in our study. More recently, low bias has been found in PFP measurements when the ambient humidity is  
124 high, based on comparisons of PFP measurements with data from in-situ analyzers at tall towers. We are working to  
125 understand and quantify this bias, and for this study we have derived a preliminary correction factor, which shows a  
126 linear trend with -1.4 ppm CO<sub>2</sub> offset per 1% above 1.7% of ambient water (mole fraction relative to whole air)  
127 content. Only ~ 4% of total aircraft measurements or ~ 12% of those below 2 km are impacted by humidity higher  
128 than 1.7%, for which we have applied corrections before data analysis. The mean correction applied is  $0.53 \pm 0.4$  (1  
129  $\sigma$ ) ppm for the impacted data.

130 The NOAA tall tower network measures CO<sub>2</sub> and other trace gases within the continental boundary layer.  
131 Continuous in-situ measurements are conducted using nondispersive infrared (NDIR) absorption sensors and cavity  
132 ring-down analyzers. The long-term stability of these systems is typically better than 0.1 ppm for CO<sub>2</sub> (Andrews et  
133 al., 2014). Most tall tower sites have more than one air intake height. In this study, continuous in-situ measurements  
134 from the highest intake are used to minimize potential influences from local sources. More information concerning  
135 the tower sites can be found at <http://www.esrl.noaa.gov/gmd/ccgg/insitu/>. For the column XCO<sub>2</sub> calculation, tower  
136 data only from 10:00-17:00 local standard time (LST) on flight days are averaged to one data point per day, as a  
137 complement to vertical profiles within the PBL.

## 138 **2.2 Site description**

139 We analyze data from 19 aircraft sites and 6 tall tower sites during 2004 to 2014 (see Table S1 for a summary of site  
140 conditions). After considering the geographic distribution of these sites in North America, we group them into eight  
141 regions for spatial comparisons (Fig. 1). The northern west (NW) and southern west (SW) regions represent the  
142 inflow area in the west coast of US, directly downwind of the Pacific Ocean at both higher elevations. The northern  
143 mid-continent (NM) region represents the boreal forest and agriculture region in north-central North America. The

144 mid-continent (MC) region represents a dry landscape due to its high elevation (above 1.5 km on average) and semi-  
145 arid climate. The mid-west (MW) region is strongly influenced by agriculture and temperate forest. The southern  
146 mid-continent (SM) represents the south-central humid temperate region, with inflow from the Gulf of Mexico  
147 during summer. The northeast (NE) region represents the temperate forest in north-east coast of U.S., which is  
148 mostly downwind of regions to the west above the PBL, and downwind of its south-west regions within the PBL.  
149 The southeast (SE) region represents the warm temperate region in the south-east coast of U.S.

### 150 **2.3 Smoothing of the reference data and column XCO<sub>2</sub> calculation**

151 We use Mauna Loa Observatory (MLO) as a reference site. MLO is located at 19.536°N, 155.576°W, and 3397 m  
152 above sea level. Carbon dioxide measurements from this site are widely used to represent background CO<sub>2</sub> in the  
153 Northern Hemisphere. For our study, a function consisting of a quadratic polynomial and four harmonics is fitted to  
154 the MLO data, adopted from the method described by Thoning et al. (1989). Residuals of the data from this function  
155 are smoothed by a low-pass filter with full-width at half-maximum in the time domain of 1.1 years. The smoothed  
156 residuals are then added back to the polynomial part of the function to produce the long-term deseasonalized trend.  
157 This trend (see Fig. 3) is subtracted from all aircraft and tall tower measurements. Also, the CarbonTracker results  
158 presented in this study are the differences relative to observed MLO deseasonalized trend. We use ‘Δ’ to represent  
159 detrended data in the following text and figures. The choice of reference site is not important for this study, since  
160 we focus on examining the relative seasonal patterns of the detrended spatial and vertical distributions of CO<sub>2</sub>  
161 instead of the total changes in CO<sub>2</sub> abundance attributed to global surface fluxes.

162 We calculate partial column average CO<sub>2</sub> dry mole fraction using tall tower and aircraft data, and the total  
163 column by adding simulations of high altitude CO<sub>2</sub> (above 330 hPa, ~ 8 km above sea level) from CarbonTracker.  
164 Since geometric height from the onboard Global Positioning System (GPS) (after 2006) or inferred from the aircraft  
165 altimeter or pressure altitude is archived with each aircraft measurement, we first convert geometric height (in  
166 meter) to pressure (in hPa) for the pressure-weighted column XCO<sub>2</sub> calculation. This conversion uses geopotential  
167 data from NOAA/NCEP North American Regional Reanalysis (NARR) (Mesinger et. al, 2004), available at  
168 <https://www.esrl.noaa.gov/psd/data/gridded/data.narr.html>, in which the geopotential is a function of latitude,  
169 longitude, pressure altitude and time. We interpolate the geopotential field vertically to retrieve pressure, and then  
170 calculate dry pressure by incorporating specific humidity data from NARR. Eventually we use a trapezoidal method  
171 to integrate over detrended vertical profiles for dry-pressure-weighted column averages. For the long-term averaged  
172 column ΔXCO<sub>2</sub> calculation, a long-term mean vertical profile is first constructed for each month by combining 11-  
173 year detrended data together and then average data in each 40 hPa vertical bin. To look at the long-term averaged  
174 total column ΔXCO<sub>2</sub> from individual aircraft sites, we combine aircraft data with upper-layer CT2015 simulations.

175 The NOAA CarbonTracker model assimilates CO<sub>2</sub> measurements from surface sampling networks and tall  
176 towers to generate global 3D fields of atmospheric CO<sub>2</sub> mole fraction. The Carbon Tracker model has evolved  
177 significantly since Peters et al. (2007). A detailed description of this model is provided in documents available at  
178 <http://carbontracker.noaa.gov>. Our study utilizes CarbonTracker results from the 2015 release (CT2015), publicly  
179 accessible at <ftp://aftp.cmdl.noaa.gov/products/carbontracker/co2/CT2015/molefractions/>. This version provides

180 CO<sub>2</sub> mole fraction over North America with 1° × 1° spatial and 3 hour temporal resolutions, which are analyzed in  
181 Sect. 3.2 and 3.3. Total column CO<sub>2</sub> calculated from CT2015 global data with 3° × 2° spatial resolution is also  
182 presented in the supporting information (SI). We have evaluated the performance of CarbonTracker in upper  
183 atmosphere (330 to 0 hPa) by comparing its simulations with in-situ measurements from 9 AirCore profiles (Karion  
184 et al., 2010) sampled in 2012-2014. AirCore is a ~150 m stainless steel tube that utilizes changes in ambient  
185 pressure for passive sampling of the vertical profile. The tube is carried to high altitude by balloon and it collects a  
186 continuous sample as it descends. It is then measured by an analyzer after it is recovered. More information about  
187 AirCore system can also be found at <https://www.esrl.noaa.gov/gmd/ccgg/aircore/>. All 9 AirCore profiles are taken  
188 near SGP and CAR sites. Figure 2 shows examples of AirCore profiles compared with CT2015 in the upper  
189 atmosphere, which demonstrates good agreement. We also compare partial column (330 to 0 hPa) averages from the  
190 9 AirCore profiles and CT2015. Results from CT2015 agree generally well with AirCore, with difference ranging  
191 from 0.03 to 1.22 ppm (mean value equals 0.66 ppm), which suggests that CT2015 may have a high bias that could  
192 contribute to  $0.66 \times 1/3 = 0.22$  ppm overestimate on average to the total column average. However, AirCore is in the  
193 process of rigorous evaluation, the differences between AirCore and CT2015 are not well characterized yet since we  
194 only have a limited amount of AirCore data. It is unclear whether the potential bias of CT2015 in this partial column  
195 is dependent on time or sampling location. Adding a constant bias correction to all regions will not change the  
196 spatial gradients that we focus on in this study. Thus no correction is applied when using CT2015 simulations to  
197 represent the upper 1/3 of the total column. For uncertainty estimates, we use a ‘bootstrap’ method that uses random  
198 resampling of individual vertical profiles with restitution (low bias, high humidity was corrected), with 100 Monte  
199 Carlo runs for each column average calculation. Uncertainty is then defined as one standard deviation of the 100  
200 Monte Carlo results.

## 201 **3 Results and Discussions**

### 202 **3.1 Seasonal patterns and spatial gradients**

203 Typically one aircraft profile contains measurements at 12 different altitudes. Column  $\Delta XCO_2$  can be computed for  
204 each profile using the method described in Sect. 2.3 (Fig. S1). Our aircraft and CT2015 based column CO<sub>2</sub> at SGP  
205 and LEF sites shows reasonable agreements with TCCON data retrieved at Lamont and Park Falls site  
206 (Washenfelder et al., 2006; Wunch et al, 2009, 2011), respectively (Fig. S2). Figure 3 shows aircraft (at all altitudes)  
207 and tower data (daily averages for 10:00-17:00 LST data) from all sites used in this study. Aircraft data above 2 km  
208 exhibit much smaller seasonal variations than the full dataset, because the variations are mainly driven by CO<sub>2</sub>  
209 sources and sinks near Earth’s surface. CO<sub>2</sub> mole fraction is enhanced in the shallow wintertime PBL primarily due  
210 to reduced plant photosynthesis and ecosystem respiration combined with slightly increased fossil fuel emissions.  
211 During summer the PBL is deeper, and depletions within the PBL are due to strong terrestrial uptake that dominates  
212 over emissions especially during June through August. During summer of 2010 to 2012, CO<sub>2</sub> from aircraft  
213 measurements appears higher than other years in Fig.3; however, similar characteristics are not present in tower  
214 data. This apparent difference is due to a decrease in sampling frequency at several aircraft sites that resulted in an

215 aliased picture of the full summer signals. Since we focus on climatological mean of 11 years of data in our study,  
216 this influence is eliminated by combining 11 years of data together into one “average year”.

217 To investigate the contributions of different altitudes to spatial gradients between regions, we divided all  
218 measurement data into three layers according to their sampling altitudes: below 2 km, 2 - 5km, and 5 - 8.5 km masl  
219 (Fig. 4). Smooth seasonal curves are attained from fitting data with four harmonics using the method described by  
220 Thoning et al. (1989). The peak-to-valley amplitudes of the seasonal cycles below 2 km are the largest among the  
221 three layers for most regions, with a minimum of 10.3 ppm in SM and a maximum of 25.0 ppm in MW. The  
222 seasonal variation amplitudes decrease to 7.7-11.5 ppm in the 2 - 5 km layer, and further decrease to 7.2-10.0 ppm in  
223 the 5 - 8.5 km layer. We also observe that the seasonal cycle drawdown occurs later in the layers above 2 km (see  
224 Fig. S3, which provides similar information as Fig. 4, but seasonal curves from different vertical layers are grouped  
225 by regions to facilitate comparisons of the phases of seasonal cycles). The seasonal CO<sub>2</sub> drawdown below 2 km is  
226 mainly influenced by terrestrial photosynthesis and gradients are due to local to regional fluxes, with an earlier onset  
227 of drawdown in southern regions than in northern regions. The seasonal cycle aloft is damped and lagged compared  
228 to the PBL, with influences from throughout the Northern Hemisphere and with spatial gradients likely driven by  
229 large-scale transport. The NW, SW, SM, and SE inflow regions have significant delays of more than one month in  
230 the 2 - 5 km layer compared with the surface layer, which is likely due to the delayed phase of the seasonal cycle in  
231 well-mixed air coming from the oceans. Vertical homogeneity of air over ocean was observed during the HIAPER  
232 Pole-to-Pole Observations (HIPPO) aircraft campaign (Wofsy et al., 2011; Frankenberg et al., 2016). As air masses  
233 are transported further inland, we observe reduced discrepancies of the timing of CO<sub>2</sub> drawdown between surface  
234 and upper layer air (2-5 km), which may be associated with the increased influence of the land surface in the mid-  
235 troposphere due to strong convection over land. CO<sub>2</sub> drawdown in the 5 - 8.5 km layers also occurs later than in the  
236 2 - 5 km layers in most regions; however, differences between these two layers are small. The declining amplitude  
237 and delayed phase of the seasonal cycle with altitude have been noted often (e.g., Tanaka et al., 1983; Ramonet et  
238 al., 2002; Gerbig et al., 2003, Sweeney et al. 2015). It demonstrates that there is lot of important information in the  
239 vertical profile that is diminished in observations of the total column.

240 We find that the largest horizontal spatial gradients between regions occur below 2 km during summer time  
241 (Fig. 4), with a maximum difference of ~15.5 ppm between MW and SM. SM and SW exhibit less pronounced  
242 seasonal cycles, which is likely associated with air masses from the Gulf of Mexico and the Pacific Ocean,  
243 respectively, whereas MW exhibits a deep summer drawdown (amplitude in seasonal cycles) partially as a result of  
244 strong regional forest and crop uptake. Crevoisier et al. (2010) estimated the surface flux over North America using  
245 vertical CO<sub>2</sub> measurements and average wind vectors, and reported that annually averaged land carbon fluxes in the  
246 western (including SW region) and southern regions (including SM region) were neutral. The SE region also  
247 demonstrates a less pronounced seasonal cycle with higher summertime levels compared with other northern  
248 regions, which may be due to the sea-breeze influence in summer within PBL. In wintertime, CO<sub>2</sub> levels in NE and  
249 MW are higher than in other regions, which result from regional fossil fuel and terrestrial biogenic emissions  
250 combined with transport from the west and south.

251 Higher altitude data (above 2 km) exhibit only small spatial gradients. In the 2 - 5 km layer, the largest gradient  
252 is 4 ppm in summer (Fig. 4b). It further decreases to less than 3 ppm in the 5 - 8.5 km layer (Fig. 4c). Figure 4d  
253 shows modeled CO<sub>2</sub> mole fractions from CT2015 for the upper troposphere and above (330 hPa to 0 hPa), which are  
254 used to fill in above the aircraft profiles for calculation of total column  $\Delta XCO_2$ . Spatial gradients in this layer are  
255 less than 0.5 ppm, suggesting that the top third of the total column has little contribution to the spatial gradients of  
256 the total column.

### 257 **3.2 Long-term mean vertical profiles**

258 To investigate the mean spatial gradients, we first calculate the long-term mean monthly vertical profiles as  
259 described in Sect. 2.3. In addition, each tower serves as one additional layer in the mean profile. The long-term  
260 mean tower data generally fit well in the vertical profiles from measurements of aircraft samples (Fig. 5 and Fig. 6),  
261 suggesting that the biases described in Sect. 2.1 above do not significantly affect the long-term mean. To attain  
262 profiles of the entire atmospheric column, upper layers (330 to 0 hPa) are filled in by CT2015, and the lowest data  
263 point of the measured profile is extended to ground level, defined by the mean surface elevation in that region.

264 Figure 5 presents two examples of long term mean profiles with data variability, which is the one standard  
265 deviation for each 40 hPa bin of aircraft data or for all flight-day tower data. Variability as large as 20 ppm is seen  
266 within the PBL in the MW region in summer, which is due to strong and heterogeneous surface vegetation uptake  
267 and ecosystem respiration combined with day-to-day changes in wind direction. All long-term mean monthly  
268 vertical profiles are presented in Fig. 6, which shows the mean temporal and vertical variability of CO<sub>2</sub> in each  
269 season, and further demonstrates the vertical propagation of seasonal CO<sub>2</sub> due to changes of surface flux. In  
270 wintertime, monotonic decrease of CO<sub>2</sub> with altitude can be observed from all regions, in which high PBL CO<sub>2</sub> is  
271 mainly driven by surface emissions and reduced vertical mixing (Denning et al., 1998; Stephens et al., 2007).  
272 Surface CO<sub>2</sub> decreases dramatically in the growing season in those regions influenced by high plant activity, such as  
273 NM and MW regions. For the summer vertical profiles in NE and SE region (east coast of the U.S.), the CO<sub>2</sub> mixing  
274 ratio is elevated in the layer under 900 hPa followed by significant decreases in upper layers until 750 hPa, and then  
275 increases with altitude until tropopause (Fig. 6). This is likely a result of sea breeze influence. Lower-troposphere air  
276 from the sea, lacking terrestrial uptake of CO<sub>2</sub>, typically has higher CO<sub>2</sub> in summer compared with inland air.  
277 Polluted air previously advected offshore can be brought back along with sea breeze. Without significant vertical  
278 mixing over the marine surface, high levels of pollutants remain in those air masses. The convergence of sea breeze  
279 with prevailing wind moving offshore may create a period with a stalled frontal structure that can aggregate air  
280 pollutants (Banta et al., 2005). The convective internal boundary layer structure of the sea breeze system can  
281 significantly reduce mixing height (Miller et al., 2003), and also induces higher CO<sub>2</sub> levels. When the sea breeze is  
282 not dominant, air advected from southwest and west (the land) can also bring in polluted air with high CO<sub>2</sub> since this  
283 region is downwind of continental U.S. emissions (Miller et al., 2012).



### 3.3 Partial column $\Delta XCO_2$ and total column $\Delta XCO_2$

Seasonal variations of monthly averaged partial column  $\Delta XCO_2$  (below 330 hPa) demonstrate maximum values in April and minimum values in August or September (Fig. 7a). The largest amplitude appears in NM, with peak-to-valley difference up to 13.5 ppm. SW, SM, SE, and MC have similar amplitudes of 7-8 ppm, smaller than the other three regions. To evaluate the performance of CT2015 on column  $\Delta XCO_2$ , CT2015 results are sampled to match the latitude, longitude, altitude and time of actual measurements (CarbonTracker team, 2016). Note that aircraft profiles are not assimilated in CT2015, so aircraft data are independent of the CT2015 data assimilation. Figure 7b shows monthly partial columns of  $\Delta XCO_2$  calculated from CT2015, which demonstrate good agreement with results from measurements. Only small seasonal biases exist in CT2015, with high bias occurring mostly in spring and early summer and low bias in September and October (Fig. S4). The overall differences of monthly partial column  $\Delta XCO_2$  (CT2015 - measurements) mainly fall in the range of -0.64 ppm (5<sup>th</sup> percentile) to 0.84 ppm (95<sup>th</sup> percentile) with a mean difference of 0.13 ppm. These differences are of similar magnitude to the uncertainties of partial column  $\Delta XCO_2$  calculated from the measurements (Fig. S5). It is clear that CT2015 captures the long-term mean variations of both phase and amplitude of partial column  $XCO_2$  reasonably well when compared with well-calibrated measurements across North America.

Total column  $\Delta XCO_2$  is presented in Fig. 7c. In regions NW, NM, NE, and MW, seasonal variations of total column  $\Delta XCO_2$  are very similar in both phase and amplitude (8-9 ppm peak to valley). For SW, SM, SE, and MC, amplitudes are ~5.5 ppm. The smallest spatial gradients occur during May and October, which result in maximum differences among all regions of only 0.9 and 0.7 ppm, respectively. The largest spatial gradients occur during June, July and August, which result in maximum differences of 2.4, 4.5, and 4.1 ppm, respectively. It is interesting that the deepest seasonal drawdown is seen in region NM, not in region MW that encompasses the very intensive agricultural activities in the U.S. mid-west, which suggest the possibility of strong upwind influence in the NM region. Transported signals have significant influences on total column  $CO_2$ . The summer total column  $\Delta XCO_2$ , represented by the June to August average from CT2015, has a magnitude that is similar to observations with differences no more than 1 ppm (Fig. 8). Based on the seasonal patterns of total column  $\Delta XCO_2$  (Fig. 7c) and the summer column  $\Delta XCO_2$  (Fig. 8), we can separate the eight regions into two groups. The group with NW, NM, NE, and MW, has ~3 ppm stronger drawdown (larger amplitude) than the group with SW, SM, SE, and MC. For winter total column  $\Delta XCO_2$  (December to February average), the maximum spatial difference is only 1.6 ppm, with the highest total column  $\Delta XCO_2$  of 1.2 ppm in NE and the lowest value of -0.3 ppm in MC.

### 3.4 Influence of large scale circulation

Figure 9 shows long-term mean summer column  $\Delta XCO_2$  calculated from CT2015, together with full column  $\Delta XCO_2$  from individual aircraft sites. Note that some aircraft sites have less than 11 years of data that CT2015 shows in Fig. 9, only aircraft sites with more than 6 years of data are presented, the actual values are provided in Table S2. The fact that total column  $\Delta XCO_2$  from CT2015 agrees well with aircraft sites supports the performance of CT2015 on a long-term average basis. The observations show a similar summer spatial pattern, with lower

column  $\Delta XCO_2$  in the north and northeast regions and higher column  $\Delta XCO_2$  in the south and southwest regions (Fig. 9a). Scattered hot spots of high column  $\Delta XCO_2$  associated with surface emissions from megacities, or cold spots associated with strong local uptake, are not or just barely visible in the long-term average column  $\Delta XCO_2$  map at  $1^\circ \times 1^\circ$  resolution. Instead, the wave-like pattern of column  $\Delta XCO_2$  over North America reflects the average large scale circulation. To support our hypothesis on the influence of large scale circulation, we analyze the long term mean wind pattern over North America. We can see that air masses from northwest of the continent bring in low average column  $\Delta XCO_2$ , while air masses from the south (mainly the subtropical Pacific Ocean and the Gulf of Mexico) bring in high column  $\Delta XCO_2$  (Fig. 9b). The zonal gradients over the continent, especially north of  $40^\circ$  N, also reflect long-term average wind patterns; southwest wind corresponds to higher column  $\Delta XCO_2$  over the western part of the continent until the wind direction shifts to west-northwest over the eastern part of the continent. This wind pattern matches well with the geographic division of the over/under -3 ppm areas colored in green/blue in the column  $\Delta XCO_2$  map (Fig. 9b). Figure 9c and 9d shows partial column averages for free troposphere (800-330 hPa) and lower troposphere (below 800 hPa), respectively. The free troposphere spatial gradient also demonstrates a wave-like pattern. A previous study on the total column  $CO_2$  from ground based Total Carbon Column Observation Network (TCCON) found strong correlation between the mid-latitude column  $CO_2$  and synoptic-scale variation of potential temperature ( $\theta$ , at 700 hPa), a dynamic tracer for adiabatic air transport (Keppel-Aleks et al., 2012). Thus they also propose that the variations in column  $CO_2$  are mainly driven by large-scale flux and transport. Analysis of the interannual variability of the seasonal cycle amplitudes of column  $CO_2$  in North Hemisphere has also found significant contribution of large-scale circulations to the north-south gradient (Wunch et al., 2013).

The strong drawdown over northeast North America in summer is a consequence of long-range transport of low  $CO_2$  from northeast Eurasia, in addition to regional terrestrial uptake. Sweeney et al. (2015) notes well-mixed vertical profiles (up to 8 km) of  $CO_2$ , CO,  $CH_4$ ,  $N_2O$ , and  $SF_6$  from THD, ESP and PFA (Poker Flat, Alaska;  $65.07^\circ$ ,  $-147.29^\circ$ ) sites and suggests that air coming across the Pacific was strongly influenced by Asian surface fluxes before being vertically homogenized as it passed over the Pacific Ocean. This well-mixed air forms an important boundary condition in the column  $CO_2$  of air coming into the North American continent. This was best illustrated at sites like PFA where the summertime minimum in  $CO_2$  significantly preceded maximum ecosystem uptake of  $CO_2$ , implying significant influence of transported air from lower latitude regions from Asia. We further conduct an experiment using Carbon Tracker to investigate the importance of this effect. A control run and a “masked run” are conducted for 2010-2012, in which the Eurasian boreal flux is turned on/off. The MLO  $CO_2$  trend from each model scenario is used as reference background and thus removed before total column  $\Delta XCO_2$  calculation. Figure 10 shows the results for 2012 summer, which is an average summer when compared with the 2004-2014 mean pattern (Fig. 9 and Fig. 11). The maximum north-south difference reduces to  $\sim 2.5$  ppm after we turn off the Eurasian boreal flux, compared with  $\sim 5$  ppm from the control run. In both control and masked scenarios, the free troposphere partial  $\Delta XCO_2$  demonstrates similar spatial patterns as for total column  $\Delta XCO_2$  (Fig. S6). This result combined with results from Sweeney et al. (2015) demonstrates that the transport of low  $CO_2$  resulting from large summertime Eurasian boreal uptake has a large contribution on the overall summer total column  $CO_2$  decrease in North America.

### 3.5 A comparison with apparent gradients over Europe

Figure 11 shows the climatological June - August mean modeled global column  $\Delta XCO_2$  map in  $3^\circ \times 2^\circ$  spatial resolution, which presents smooth wave-like patterns. Reuter et al. (2014) use SCIAMACHY and GOSAT satellite retrievals of column  $CO_2$  and inverse modelling to infer a very large net  $CO_2$  uptake flux over European region. Column  $\Delta XCO_2$  from CT2015 (Fig. 11) exhibits a drastically different summer spatial pattern over Europe compared with the eight year mean (2003-2010) June through August satellite retrievals presented by Reuter et al. (2014, their Fig. 2a). The spatial gradient from CT2015 results in a maximum 3-4 ppm difference and a gradual pattern, instead of as much as 6 ppm from satellite retrievals. There is no sign of  $XCO_2$  hot spots from surface emissions or removals in the CT2015 spatial pattern over Europe (Fig. 11), in contrast to several hot spots that are apparent from the 8-year averaged SCIAMACHY satellite retrievals over Ireland, U.K., Belgium, Netherland, north of Germany, and south of Sweden, and low spots over the Ukraine and Kazakhstan (Reuter et al., 2014). This SCIAMACHY retrieval pattern contradicts our understanding of the significant influence of large-scale transport on column  $\Delta XCO_2$ . Although the NOAA/ESRL CT2015 (<https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2015/>) assimilates fewer observations over Europe than Carbon Tracker Europe (<http://www.carbontracker.eu/>), both models produced similar fluxes over the European region (see both websites for detailed fluxes). The  $3^\circ \times 2^\circ$  grid from CT2015 is not likely responsible for a much smoother pattern for Carbon Tracker, compared with the  $2^\circ \times 2^\circ$  grid from satellite retrievals (Reuter et al., 2014). The North America region on the  $3^\circ \times 2^\circ$  grid in Fig. 11 shows similar pattern as the  $1^\circ \times 1^\circ$  grid in Fig. 9, with similar spatial difference of  $\sim 5$  ppm. A smoother spatial distribution should be expected in Europe for the long-term mean column  $XCO_2$  (Fig. 11) due to the influences of dominating west and southwest winds in summer. We have also evaluated the importance of sampling bias by sampling CT2015 at the same latitude/longitude/hour (within 1 hour) as in SCIAMACHY BESD (v02.00.08) data (Reuter et al., 2011). The 8-year mean pattern shows much smaller gradients (3-4 ppm maximum) without significant hot/cold spots at the locations of SCIAMACHY (Fig. S7). Although sampling biases contribute to the unphysical column  $XCO_2$  spatial pattern from SCIAMACHY, they cannot explain the large gradients. When we compare CT2015 directly with SCIAMACHY BESD data, we find up to  $\sim 3$  ppm differences over North America and Europe (Fig. S8). Since CT2015 compares well with calibrated data over North America, we are deeply skeptical about any sources/sinks inferred from the SCIAMACHY BESD data. A recent study (Feng et al., 2016) using inverse modeling suggests that satellite retrievals outside the immediate European region and a small bias of only 0.5 ppm were sufficient to produce the apparent large carbon sink in the study of Reuter et al. (2014). This is expected from elementary mass balance considerations as in Sec.1 above. Spatial gradients are the fundamental signals to infer regional fluxes. Since spatial gradients from CT2015 are realistic, boreal fluxes inferred by CT2015, should be more trustworthy than fluxes estimated based on unrealistic spatial pattern. The European carbon sink is still inconclusive; the discrepancies among different methods and results are further discussed by Reuter et al. (2017).

## 4 Conclusion

Aircraft and tall tower measurements from the NOAA GGGRN provide detailed information describing the long-term average temporal and spatial variations of CO<sub>2</sub> in the PBL and the free troposphere. These data provide valuable constraints for evaluating model simulations and satellite retrievals. Seasonal cycle peak-to-peak amplitudes of CO<sub>2</sub> are largest below 2 km, where those maximum values are about twice those in the vertical layers above, indicating that most of the information on surface sources and sinks resides in the continental PBL. Large spatial gradients of CO<sub>2</sub> over North America are observed below 2 km during summer (with maximum difference of ~15.5 ppm between MW and SM), while higher altitude data (above 2 km) have much smaller contributions to spatial gradients, with a maximum difference of 4 ppm. The spatial differences of CO<sub>2</sub> in the upper troposphere and above (330 hPa to 0 hPa) are less than 0.5 ppm, according to CT2015. Comparison with Aircore measurements shows CT2015 performs well simulating upper tropospheric and lower stratospheric patterns.

Our long-term mean vertical profiles show that tower data agree well with aircraft data at similar vertical levels. Partial column  $\Delta XCO_2$  was calculated from the long-term mean vertical profiles. By comparing the partial column  $\Delta XCO_2$  from measurements with those from CT2015, we verify that CT2015 captures the long-term mean patterns of both phase and amplitude of partial  $\Delta XCO_2$ .

Large spatial gradients of  $\Delta XCO_2$  only appeared in summer, during which time the north and northeast regions had ~3 ppm stronger drawdowns than the south and southwest regions. Scattered hot spots of high column  $\Delta XCO_2$  associated with surface emissions from megacities, or cold spots associated with strong local uptake, are not or just barely visible in the long-term average column  $\Delta XCO_2$ . Instead, the wave-like pattern of column  $\Delta XCO_2$  over North America matches well with the average large scale circulation. A CarbonTracker experiment to investigate the impact of Eurasian long-range transport suggests that the large summer time Eurasian boreal flux alone contributes about half of the north-south column  $\Delta XCO_2$  gradient across North America. Considering the transported signals from other upwind regions, including northern Canada, we expect that the transported signals have the overall largest contribution to the total column  $\Delta XCO_2$  spatial gradient.

### Author contributions

Xin Lan was responsible for study design, data analysis, and manuscript writing. Pieter Tans was responsible for study design, data analysis, and manuscript improvement. Colm Sweeney and Arlyn Andrews provided measurement data and improved manuscript. Andrew Jacobson provided modelled data and improved manuscript. Edward Dlugokencky analyzed measurements and ensured data quality, and improved manuscript. Jonathan Kofler conducted tower measurements and improved manuscript. Molly Crotwell, Patricia Lang, and Sonja Wolter analyzed measurements and ensured data quality. Kirk Thoning provided data smoothing method.

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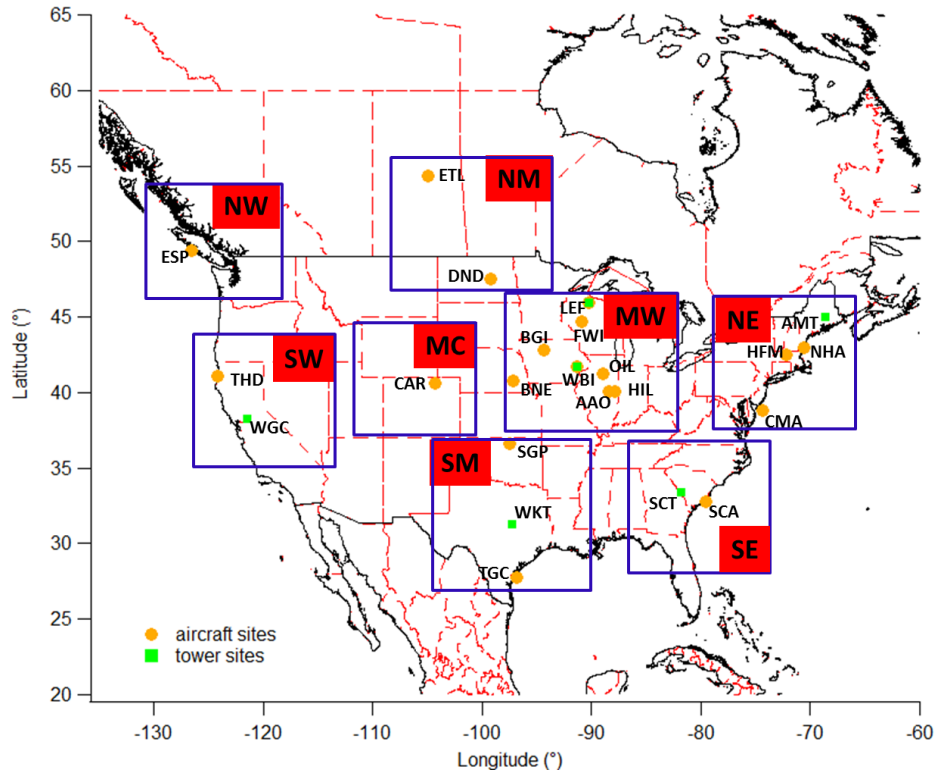
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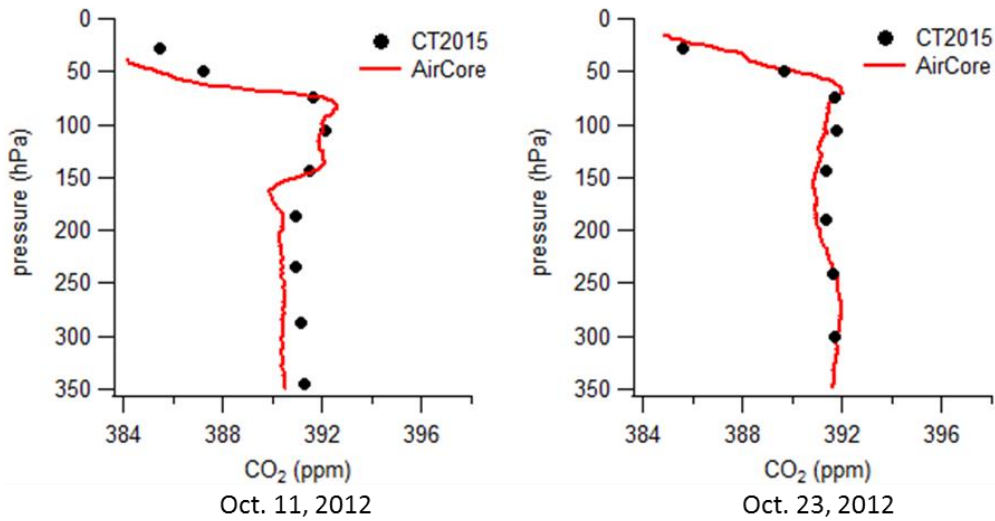


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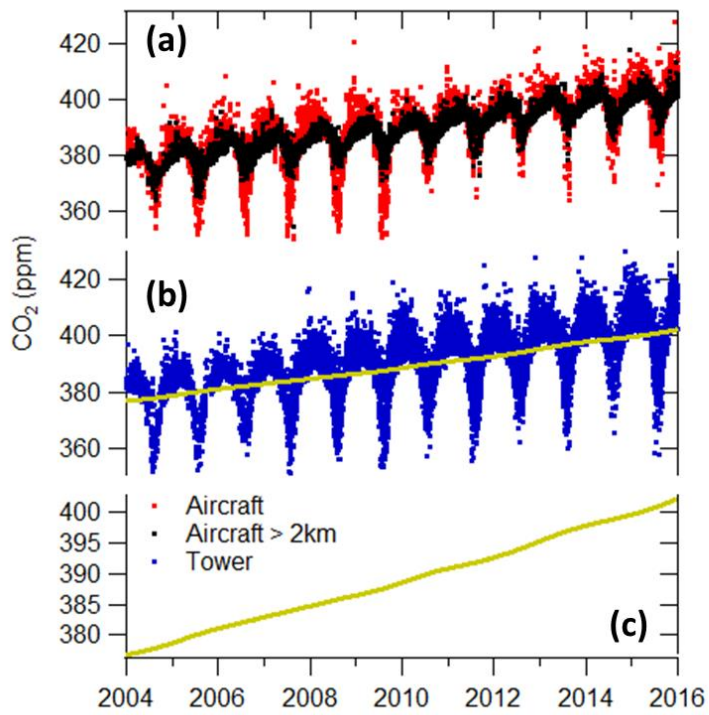
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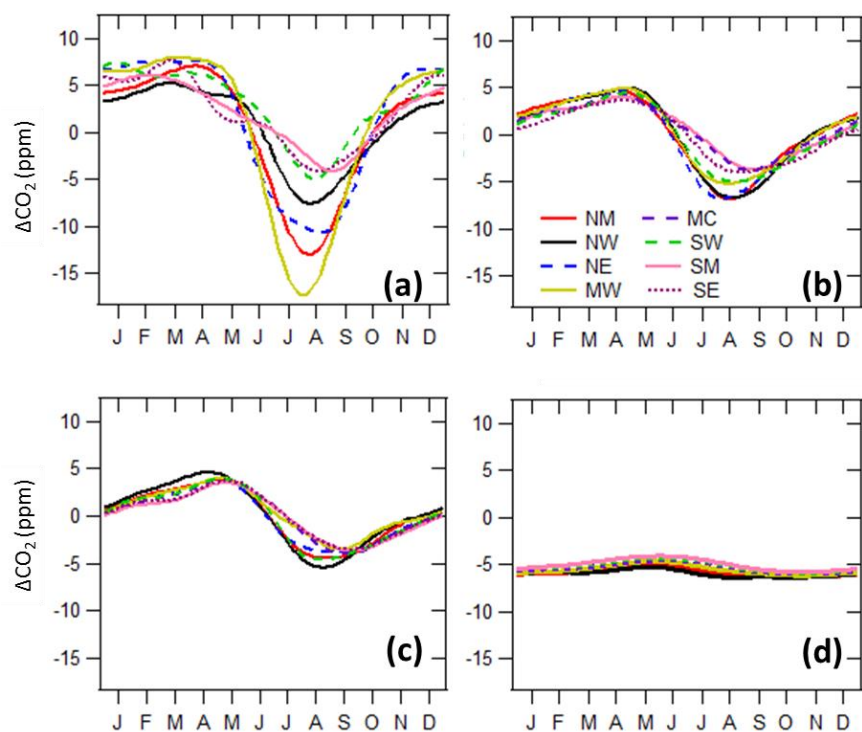
**Fig. 1.** Aircraft, tall tower, and high elevation/tower sites in the NOAA GGGRN. The eight boxes define regions that are further discussed for spatial pattern comparison.



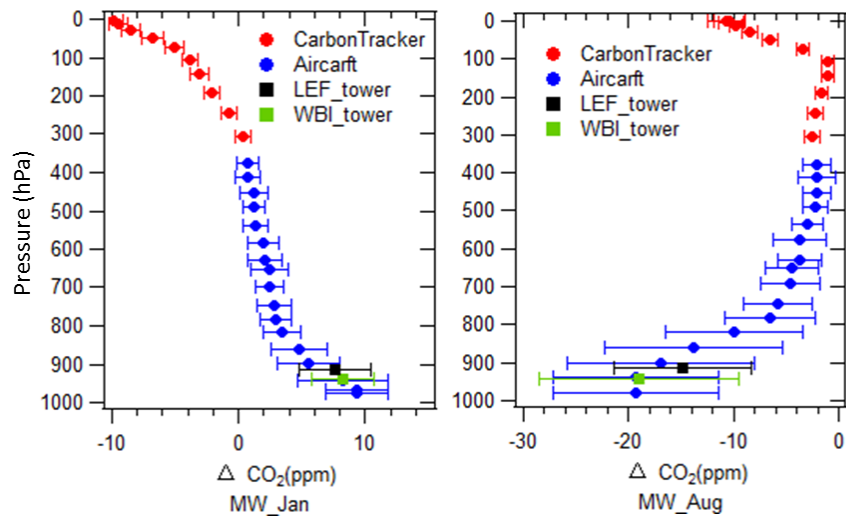
**Fig. 2.** Carbon Tracker (CT2015) simulations compared with AirCore in-situ measurements in upper atmosphere. AirCore profiles in the left and right panels are sampled near CAR and SGP, respectively.



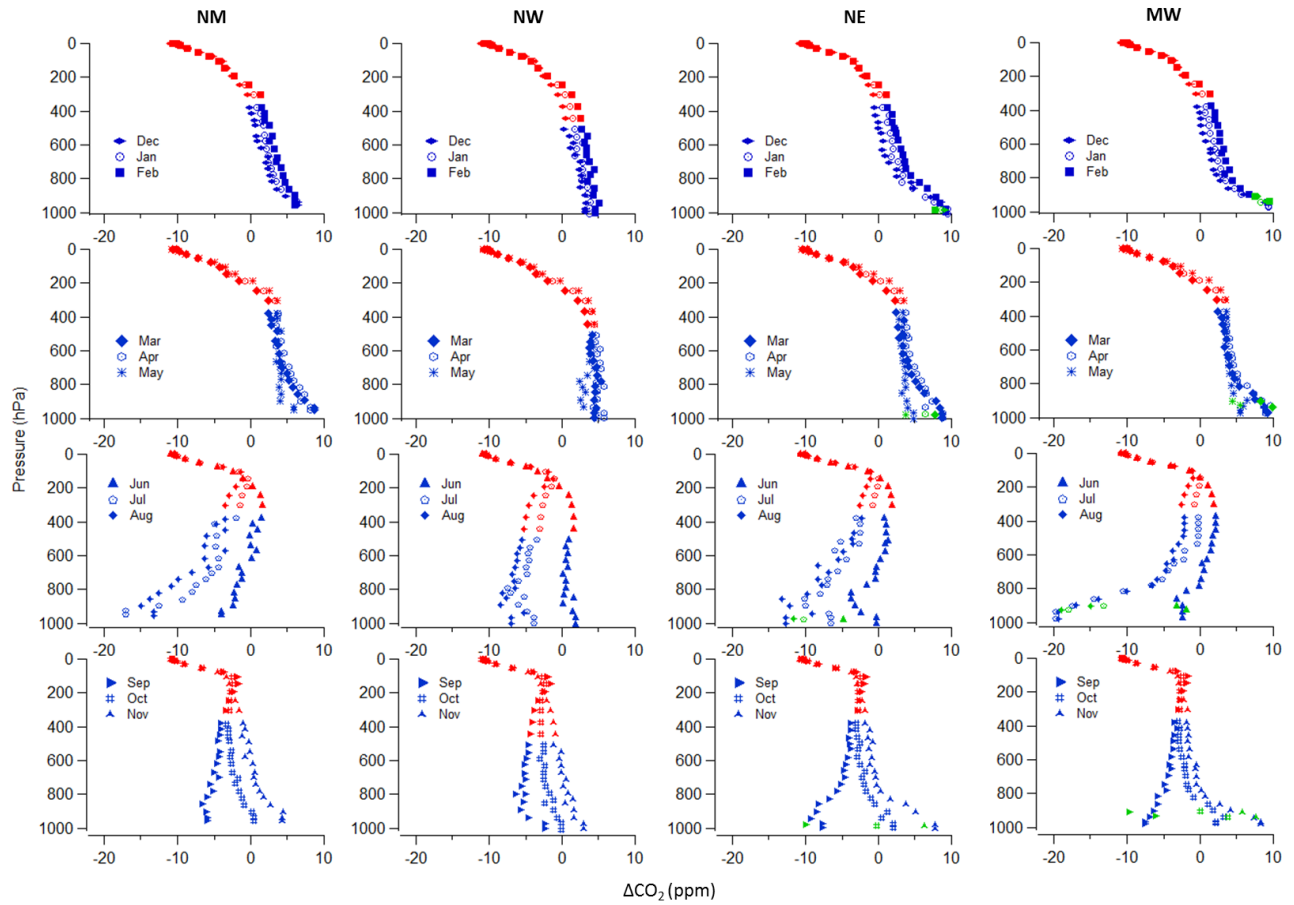
**Fig. 3.** CO<sub>2</sub> observations from aircraft (a) and towers (b). The yellow line in (b) illustrates the deseasonalized trend at Mauna Loa (MLO), same as in (c), in which y-axis is expanded.



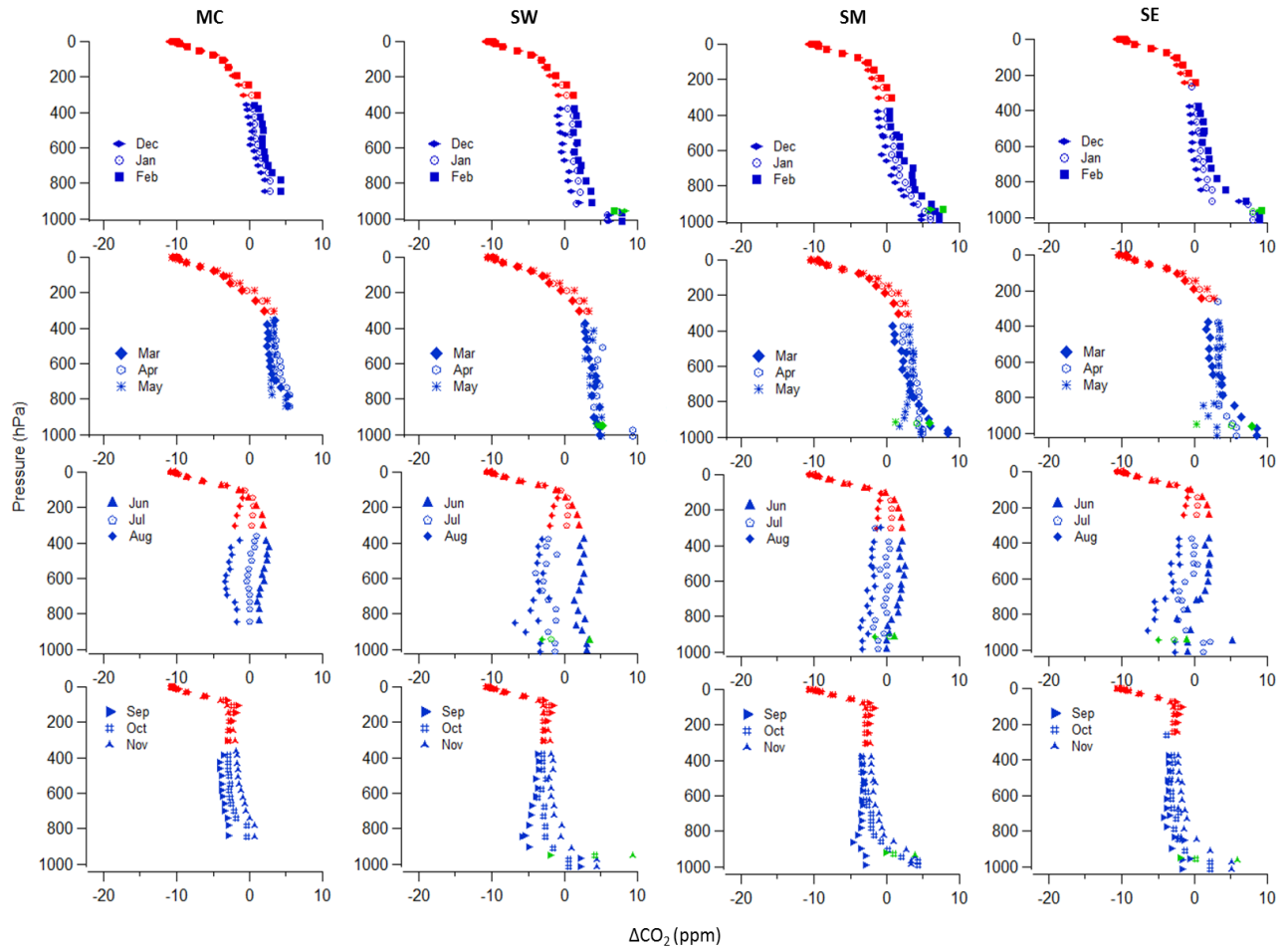
**Fig. 4.** Multi-year (2004-2014) average smooth seasonal curves of  $\text{CO}_2$  relative to the long-term de-seasonalized trend at Mauna Loa for different vertical layers: (a). Aircraft and tower data under 2 km, MC is not presented because only limited data were available due to high surface elevations ( $>1.5$  km on average) in this region; (b). Aircraft data from 2 - 5 km; (c). Aircraft data from 5 - 8.5 km; (d). CT2015 model results for layers above 330 hPa ( $\sim 8.5$  km) to 0 hPa ( $\sim 80$  km).



**Fig. 5.** Long-term mean (2004-2014) average vertical profiles in January (left panel) and August (right panel) in region MW. Error bar shows one standard deviation.

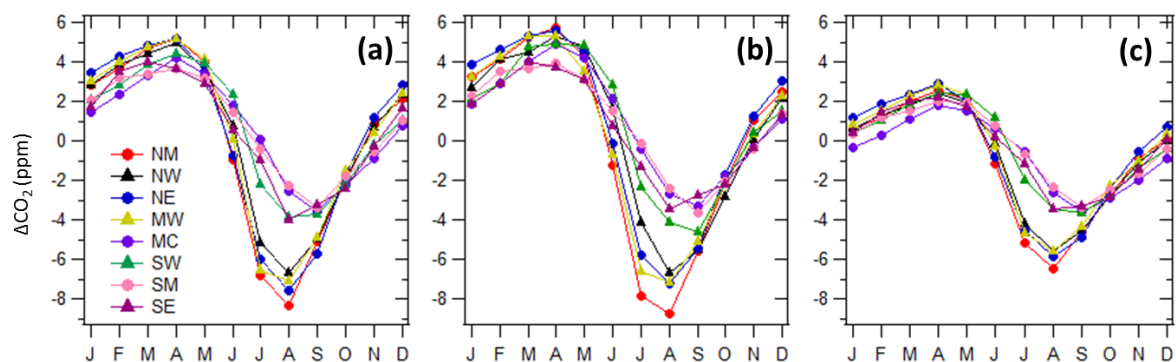


**Fig. 6a.** Long-term mean (2004-2014) monthly vertical profiles in NM, NW, NE, MW (by column, from left to right). Blue points were calculated from observations, red points were calculated from CT2015, and green points were calculated from tower data.

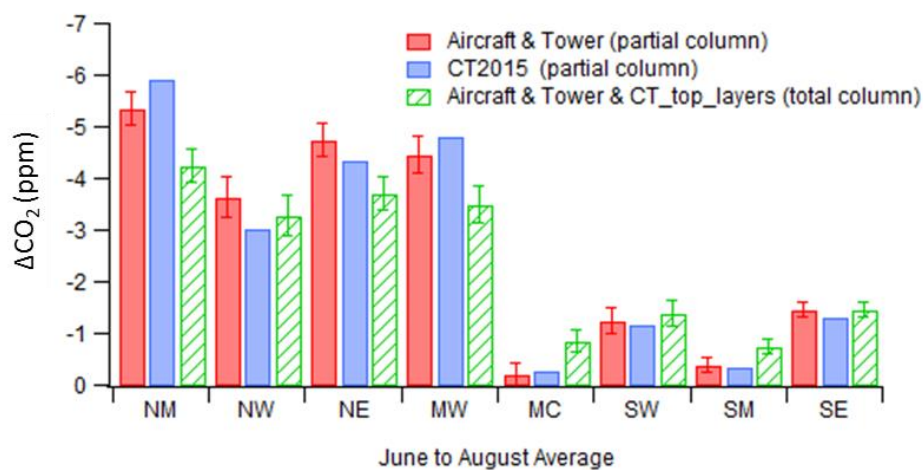


**Fig. 6b.** Long-term mean (2004-2014) monthly vertical profiles in MC, SW, SM, SE (by column, from left to right). Blue points were calculated from observations, red points were calculated from CT2015, and green points were calculated from tower data.

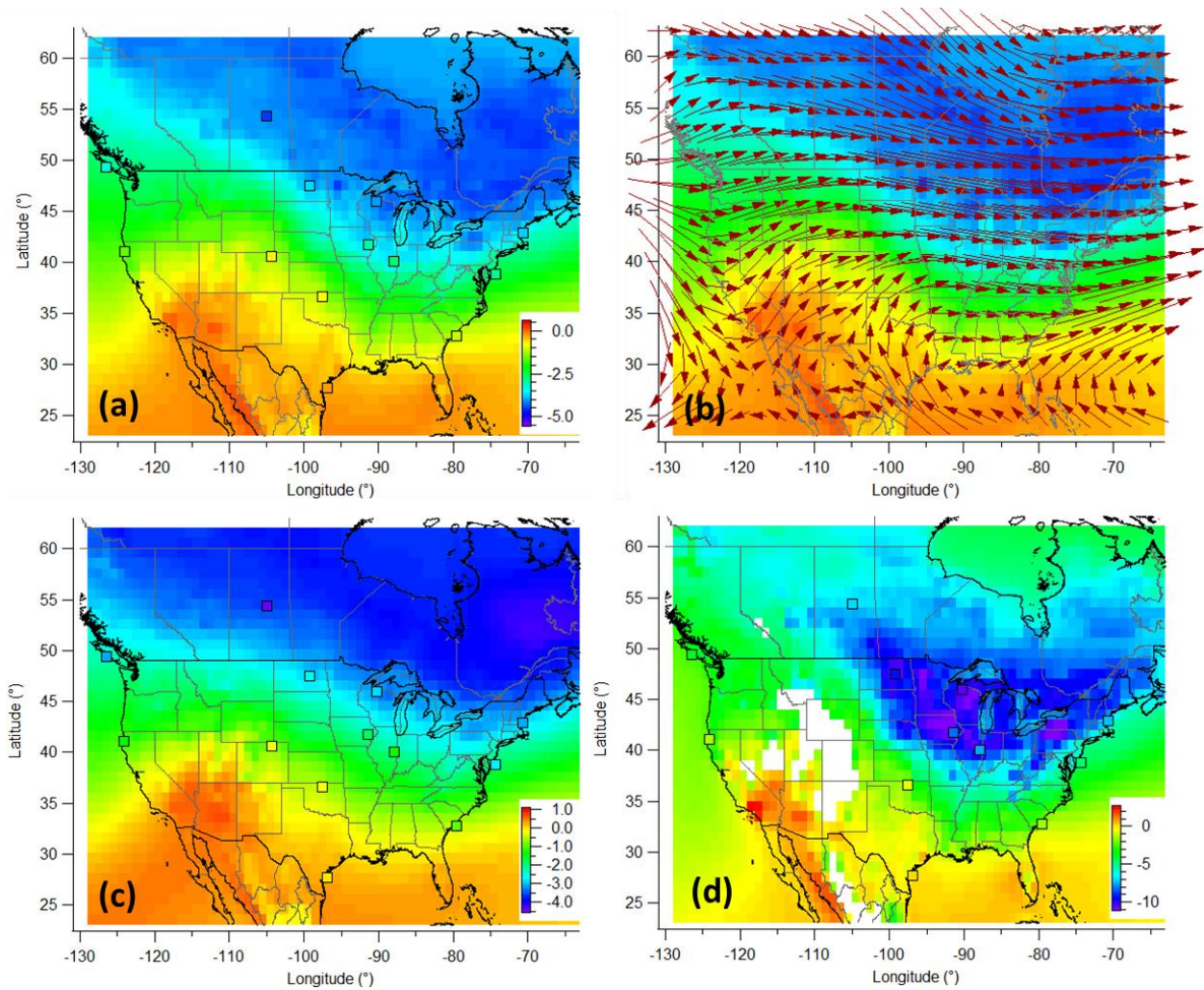




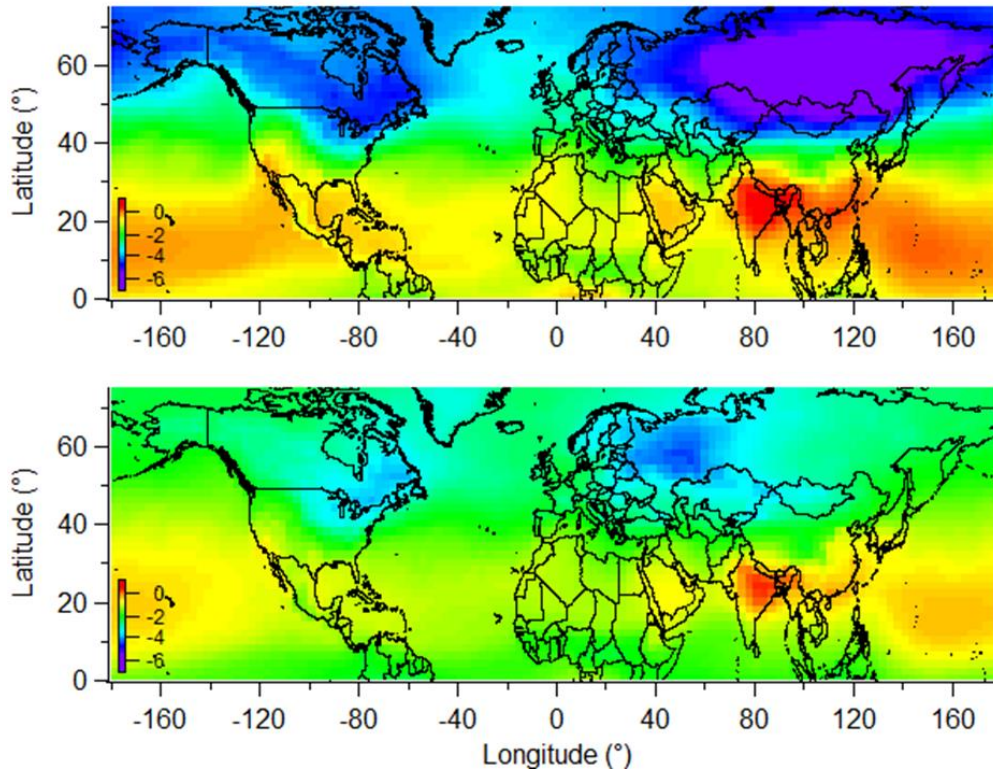
**Fig. 7.** (a). Partial column  $\Delta XCO_2$  calculated from aircraft and tower data; (b). Partial column  $\Delta XCO_2$  calculated from CT2015; (c). Total column  $\Delta XCO_2$  calculated from aircraft and tower data and including the top layer data from CT2015.



**Fig. 8.** Long-term mean (2004-2014) June to August partial and total column  $\Delta XCO_2$ . Error bars represent one standard deviation from the bootstrap uncertainty calculation (see Sect. 2.3).

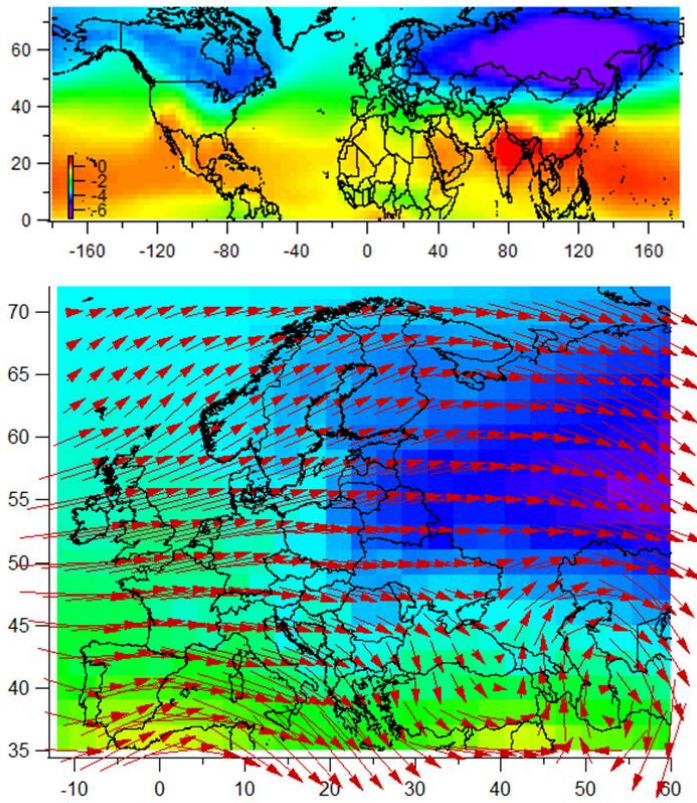


**Fig. 9.** Long-term mean (2004-2014) June-August total column  $\Delta XCO_2$  (in ppm) from CT2015 in  $1^\circ \times 1^\circ$  spatial resolution with total column  $\Delta XCO_2$  for 13 individual aircraft sites in squares (a), and CT2015 column  $\Delta XCO_2$  overlaid with pressure-weighted (1000 hPa to 500 hPa) mean wind vectors for the same period (b). (c) and (d) are similar as (a), except for free troposphere (800 to 330 hPa) and lower troposphere (below 800 hPa), respectively. Note the different color scales.



**Fig. 10.** Total column  $\Delta XCO_2$  (in ppm) from Carbon Tracker control (top panel) and masked (bottom panel, Eurasian boreal flux is masked) runs for 2012 June-August ( $3^\circ \times 2^\circ$  spatial resolution). MLO trend from each individual scenario is removed before the  $\Delta XCO_2$  calculation. Same color scale is used as in Fig. 9a. Partial column  $\Delta XCO_2$  patterns for free troposphere (800 to 330 hPa) and lower troposphere (below 800 hPa) are provided in SI.





**Fig. 11.** Long-term mean (2004-2014) June - August total column  $\Delta XCO_2$  (in ppm) from CT2015 (top panel) in  $3^\circ \times 2^\circ$  spatial resolution, and detail for Europe overlaid with pressure-weighted (1000 hPa to 500 hPa) mean wind vectors for the same period (bottom panel). The color scale is the same as in Fig. 9a, which is scaled to reflect 6 ppm difference of  $XCO_2$  to compare with satellite retrievals from Reuter et al. (their Fig. 2a, 2014).