Response to editor

Dear editor,

Thank you for editing our manuscript. Regarding to your comment:

1) the sampling of CT2015 CO2 fields does not take into account any clear sky criterion (only then can columns be retrieved from the satellite) except for matching the date and location. I do see still potential for a larger sampling bias. In the model the cloud cover could well be different. Also if the sampling was not made during the same hour of the day (rather than just during the same date), a bias can arise. This should be clarified and included in section 3.5 of the manuscript.

We actually sampled the CT2015 within 1 hour of the SCIAMACHY BESD data (version 02.00.08, same as in Reuter et al., 2014). We matched the latitude/longitude/hour when there's available SCIAMCHY BESD data (the columns were already retrieved, so model cloud is not a concern in this case). We will clarify this point in our revised manuscript.

2) The patterns in the CT2005 columns when sampling at SCIAMACHY locations/dates shown in the "Figure for reviewing process" are also not smooth, in fact in terms of smoothness they are not so different from those shown in Fig. 2a in Reuter et al., 2015, and they are more different from the smooth patterns in Fig. 11. This indicates a rather strong impact of a sampling bias, even when no additional criterion for cloudiness or hour of day is used.

The patterns we provided in the "Figure for reviewing process" is a 3-year average, unlike the 8-year average in Reuter et al. (2014) or the 11 year average in the Fig.11. Thus the color looks less smooth overall. However, our focuses are the gradients and the locations of strong hot/cold spots. The 3-year average already shows much smaller spatial difference (in ppm) compared with Reuter et al. (2014), and we cannot find cold/hot spots in those locations as in Reuter et al., Fig 2. To make our case stronger, we have updated the 3-year average to an 8-year average map in our revised manuscript. In this case our sampling period and data points can perfectly match those in Reuter et al., 2014, Fig. 2. The 8-year average column CO_2 shows a maximum 3-4 ppm difference and a gradual pattern, similar as our Fig.11 (see figure below). We

understand that sampling bias has played a role in the Reuter et al. (2014) Figure 2, especially over Ireland, UK, and south Sweden regions with very little amount of samples (it is surprising that these areas are included in Reuter et al., 2014 Fig. 2); however, it cannot explain the large differences and the strong hot/cold spots over the circled areas (hot spots over Belgium, Netherland, north of Germany, and low spots over the Ukraine and Kazakhstan). In addition, when we compare CT2015 at the time and place of our measured aircraft profiles with the measured data over the North America (lower sampling frequency for most aircraft sites compared with satellite), we do not see such strong biases (our Fig. 7 and Fig. 9).
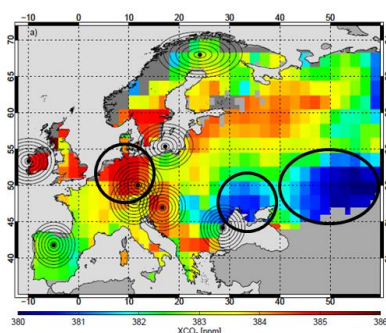
(Reuter et al., 2014, Fig. 2)
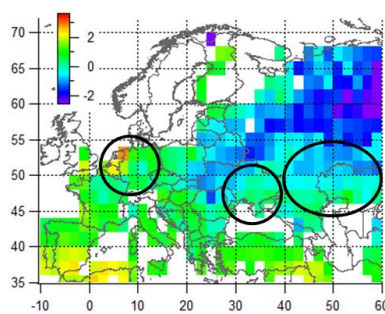
2003-2010 Jun-Aug pattern from sampling CT2015 with SCIAMACHY BESD latitude/longitude/hour. Jun-Aug mean for each year is first subtracted before putting 8-year data together and averaged within each grid (2 º × 2º). Grids with less than 24 soundings during 8 summers are excluded.
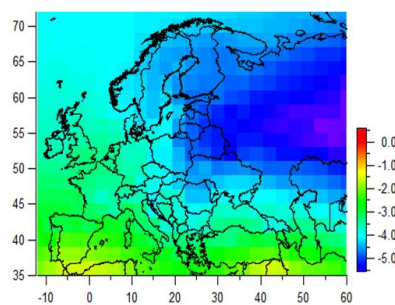
Our Fig. 11, the Mauna Loa deseasonalized trend is subtracted in this figure, but the color scale is set to reflect maximum 6 ppm difference.

Later on you state in your reply: "The reviewer seems to be confirming that, in fact, SCIAMACHY's sampling biases prevent scientists from being able to infer reliable sources/sinks." This is completely in contrast to how I interpret this reviewers comment. All the reviewer is saying is that so far you haven't been able to prove that the satellite retrievals are unphysical, as sampling biases can potentially explain this. I share this opinion, and I would think unless a sampling bias can be excluded as a reason, the language in section 3.5 needs to be toned down significantly.

We will provide the above 2003-2010 averages figure in supporting information in the revised manuscript. This figure clearly demonstrates that the sampling bias cannot explain the 6 ppm difference over Belgium, Netherland, north of Germany, versus Ukraine and Kazakhstan.

Gradients of Column CO_2 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_2) 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_2 dry mole fraction (XCO_2) in the column below ~330 hPa (~ 8 km above sea level) from NOAA's CO_2 data assimilation model, CarbonTracker (CT2015), demonstrates good agreement with those calculated from calibrated measurements on aircraft and towers. Total column XCO_2 was attained by combining modeled CO_2 above 330 hPa from CT2015 with the measurements. We find large spatial gradients of total column XCO_2 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_2 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_2 gradient across North America. Our results confirm that continental-scale total column XCO_2 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_2) from ground and airborne platforms have greatly increased our knowledge of the global carbon cycle. Observations of CO_2, 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_2 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_2 (e.g., Gerbig et al., 2003; Choi et al., 2008; Biraud et al., 2013). Routine aircraft

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36 measurements from the NOAA/ESRL GGGRN monitor the large-scale distributions of a suite of trace gases,

37 including CO_2, 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_2 measurements, such as Japan's CONTRAIL

39 (Comprehensive Observation Network for TRace gases by AIrLiner) project, which has provided valuable

40 information for CO_2 in the high troposphere and lower stratosphere (Machida et al., 2002; Machida et al., 2008).

41 Vertical profiles of atmospheric CO_2 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_2 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_2 sources and sinks are well constrained at the global scale by global mass balance, it remains

46 challenging to accurately resolve CO_2 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_2. 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_2

51 emissions from fossil fuel burning (~1.4 Pg yr^{-1}) 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^{12} m^2).

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_2 measurements have been used to evaluate ground-based total

60 column XCO_2 (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_2 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_2, 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_2 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_2 (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_2 (Inoue et al.,2016). The Orbiting Carbon Observatory-

72 2 (OCO-2) retrieval of total column XCO_2 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_2 calculated from in-situ measurements.

75 Total column XCO_2 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 XCO2 suggests that

79 they can be used to evaluate satellite retrievals of column averaged CO_2. Since aircraft profiles co-located with

80 satellite retrievals are rare, it is useful to consider the statistics of total column XCO_2 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_2 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_2 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_2 (Inoue et al., 2013).

87   Transparent and objective estimates of CO_2 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_4 and CO_2 emissions from warming permafrost in Arctic regions. Satellite retrievals of total column

91 XCO_2 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_2 over North America using well-

94 calibrated CO_2 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_2. The aircraft data enable direct analysis of column CO_2

97 characteristics, which is the fundamental step for accurate apportionment of sources and sinks. This study focuses on

98 long-term averaged column CO_2 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_2

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_2 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_2 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_2 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 ± 0.4 (1
129 σ) ppm for the impacted data.

130   The NOAA tall tower network measures CO_2 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_2 (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_2 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.

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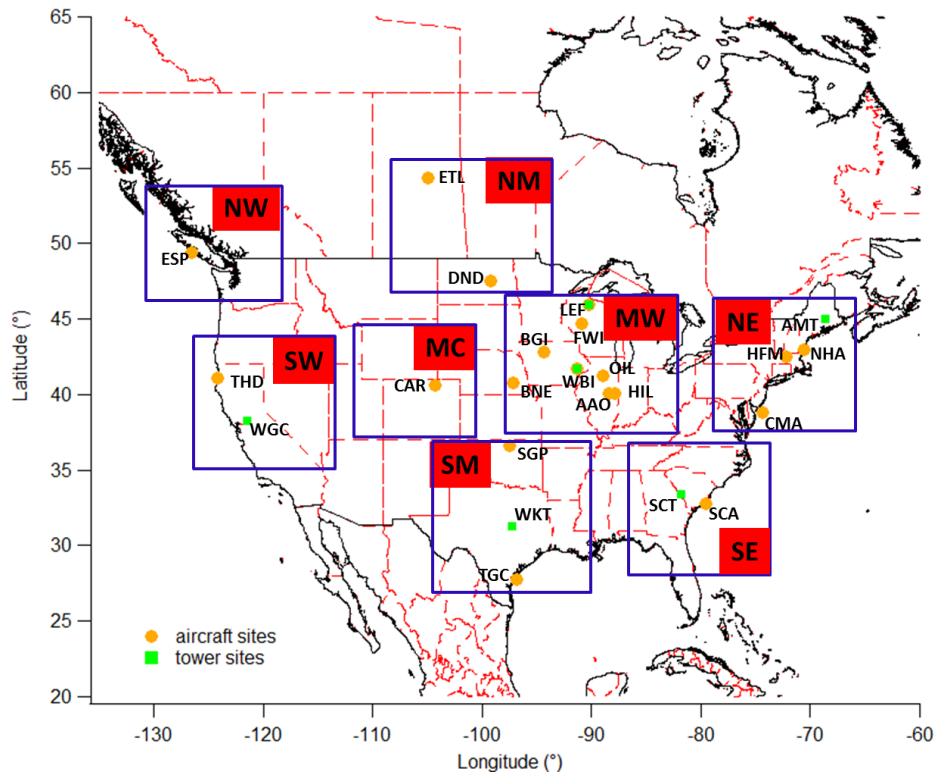
151
152 **Fig. 1.** Aircraft, tall tower, and high elevation/tower sites in the NOAA GGGRN. The eight boxes define regions
153 that are further discussed for spatial pattern comparison.

154 **2.3 Smoothing of the reference data and column XCO$_2$ calculation**

155 We use Mauna Loa Observatory (MLO) as a reference site. MLO is located at 19.536°N, 155.576°W, and 3397 m
156 above sea level. Carbon dioxide measurements from this site are widely used to represent background CO$_2$ in the
157 Northern Hemisphere. For our study, a function consisting of a quadratic polynomial and four harmonics is fitted to
158 the MLO data, adopted from the method described by Thoning et al. (1989). Residuals of the data from this function
159 are smoothed by a low-pass filter with full-width at half-maximum in the time domain of 1.1 years. The smoothed
160 residuals are then added back to the polynomial part of the function to produce the long-term deseasonalized trend.
161 This trend (see Fig. 3) is subtracted from all aircraft and tall tower measurements. Also, the CarbonTracker results
162 presented in this study are the differences relative to observed MLO deseasonalized trend. We use 'Δ' to represent
163 detrended data in the following text and figures. The choice of reference site is not important for this study, since

7

164   we focus on examining the relative seasonal patterns of the detrended spatial and vertical distributions of CO_2

165   instead of the total changes in CO_2 abundance attributed to global surface fluxes.

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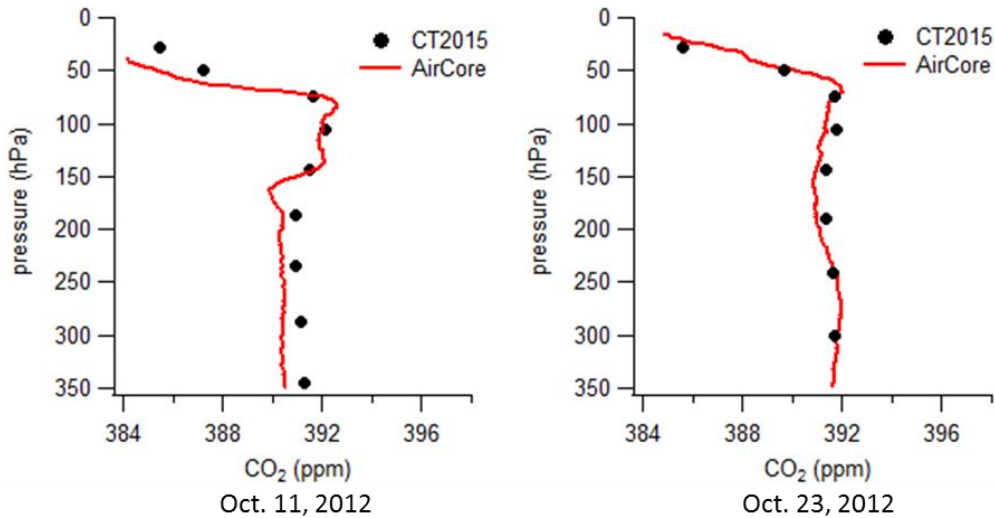
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168   **Fig. 2**. Carbon Tracker (CT2015) simulations compared with AirCore in-situ measurements in upper atmosphere.

169   AirCore profiles in the left and right panels are sampled near CAR and SGP, respectively.

170

171   We calculate partial column average CO_2 dry mole fraction using tall tower and aircraft data, and the total

172   column by adding simulations of high altitude CO_2 (above 330 hPa, ~ 8 km above sea level) from CarbonTracker.

173   Since geometric height from the onboard Global Positioning System (GPS) (after 2006) or inferred from the aircraft

174   altimeter or pressure altitude is archived with each aircraft measurement, we first convert geometric height (in

175   meter) to pressure (in hPa) for the pressure-weighted column XCO_2 calculation. This conversion uses geopotential

176   data from NOAA/NCEP North American Regional Reanalysis (NARR) (Mesinger et. al, 2004), available at

177   https://www.esrl.noaa.gov/psd/data/gridded/data.narr.html, in which the geopotential is a function of latitude,

178   longitude, pressure altitude and time. We interpolate the geopotential field vertically to retrieve pressure, and then

179   calculate dry pressure by incorporating specific humidity data from NARR. Eventually we use a trapezoidal method

180   to integrate over detrended vertical profiles for dry-pressure-weighted column averages. For the long-term averaged

181   column ΔXCO_2 calculation, a long-term mean vertical profile is first constructed for each month by combining 11-

182   year detrended data together and then average data in each 40 hPa vertical bin. To look at the long-term averaged

183   total column ΔXCO_2 from individual aircraft sites, we combine aircraft data with upper-layer CT2015 simulations.

184   The NOAA CarbonTracker model assimilates CO_2 measurements from surface sampling networks and tall

185   towers to generate global 3D fields of atmospheric CO_2 mole fraction. The Carbon Tracker model has evolved

186   significantly since Peters et al. (2007). A detailed description of this model is provided in documents available at

187   http://carbontracker.noaa.gov. Our study utilizes CarbonTracker results from the 2015 release (CT2015), publicly

188   accessible at ftp://aftp.cmdl.noaa.gov/products/carbontracker/co2/CT2015/molefractions/. This version provides

189   CO_2 mole fraction over North America with $1° \times 1°$ spatial and 3 hour temporal resolutions, which are analyzed in

190   Sect. 3.2 and 3.3. Total column CO_2 calculated from CT2015 global data with $3° \times 2°$ spatial resolution is also

191   presented in the supporting information (SI). We have evaluated the performance of CarbonTracker in upper

192   atmosphere (330 to 0 hPa) by comparing its simulations with in-situ measurements from 9 AirCore profiles (Karion

193   et al., 2010) sampled in 2012-2014. AirCore is a ~150 m stainless steel tube that utilizes changes in ambient

194   pressure for passive sampling of the vertical profile. The tube is carried to high altitude by balloon and it collects a

195   continuous sample as it descends. It is then measured by an analyzer after it is recovered. More information about

196   AirCore system can also be found at https://www.esrl.noaa.gov/gmd/ccgg/aircore/. All 9 AirCore profiles are taken

197   near SGP and CAR sites. Figure 2 shows examples of AirCore profiles compared with CT2015 in the upper

198   atmosphere, which demonstrates good agreement. We also compare partial column (330 to 0 hPa) averages from the

199   9 AirCore profiles and CT2015. Results from CT2015 agree generally well with AirCore, with difference ranging

200   from 0.03 to 1.22 ppm (mean value equals 0.66 ppm), which suggests that CT2015 may have a high bias that could

201   contribute to $0.66 \times 1/3 = 0.22$ ppm overestimate on average to the total column average. However, AirCore is in the

202   process of rigorous evaluation, the differences between AirCore and CT2015 are not well characterized yet since we

203   only have a limited amount of AirCore data. It is unclear whether the potential bias of CT2015 in this partial column

204   is dependent on time or sampling location. Adding a constant bias correction to all regions will not change the

205   spatial gradients that we focus on in this study. Thus no correction is applied when using CT2015 simulations to

206   represent the upper 1/3 of the total column. For uncertainty estimates, we use a "bootstrap'

207   method that uses random resampling of individual vertical profiles with restitution (low bias, high humidity was

208   corrected), with 100 Monte Carlo runs for each column average calculation. Uncertainty is then defined as one

209   standard deviation of the 100 Monte Carlo results.

210   **3 Results and Discussions**

211   **3.1 Seasonal patterns and spatial gradients**

212   Typically one aircraft profile contains measurements at 12 different altitudes. Column ΔXCO_2 can be computed for

213   each profile using the method described in Sect. 2.3 (Fig. S1). Our aircraft and CT2015 based column CO_2 at SGP

214   and LEF sites shows reasonable agreements with TCCON data retrieved at Lamont and Park Falls site

215   (Washenfelder et al., 2006; Wunch et al, 2009, 2011), respectively (Fig. S2). Figure 3 shows aircraft (at all altitudes)

216   and tower data (daily averages for 10:00-17:00 LST data) from all sites used in this study. Aircraft data above 2 km

217   exhibit much smaller seasonal variations than the full dataset, because the variations are mainly driven by CO_2

218   sources and sinks near Earth's surface. CO_2 mole fraction is enhanced in the shallow wintertime PBL primarily due

219   to reduced plant photosynthesis and ecosystem respiration combined with slightly increased fossil fuel emissions.

220   During summer the PBL is deeper, and depletions within the PBL are due to strong terrestrial uptake that dominates

221   over emissions especially during June through August. During summer of 2010 to 2012, CO_2 from aircraft

222   measurements appears higher than other years in Fig.3; however, similar characteristics are not present in tower

223   data. This apparent difference is due to a decrease in sampling frequency at several aircraft sites that resulted in an

224 aliased picture of the full summer signals. Since we focus on climatological mean of 11 years of data in our study,

225 this influence is eliminated by combining 11 years of data together into one "average year".


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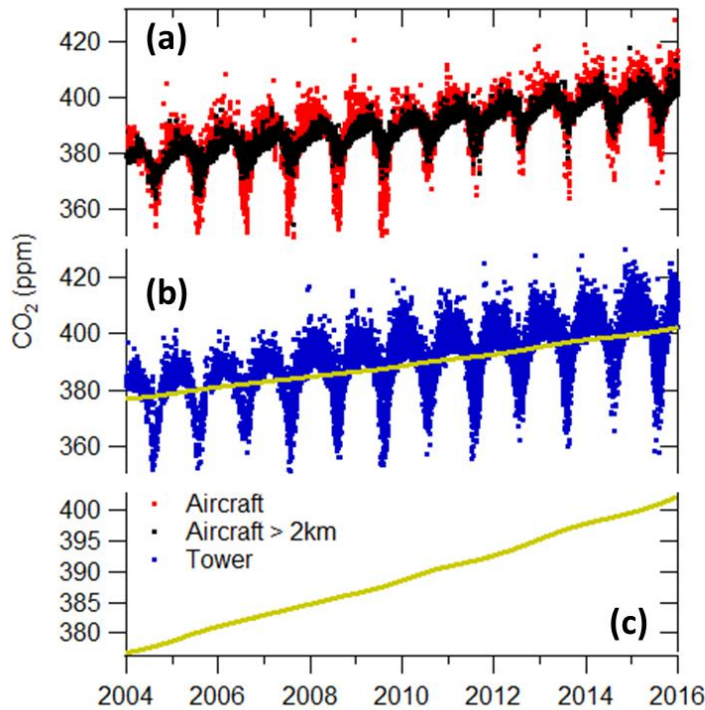
Fig. 3. CO_2 observations from aircraft (a) and towers (b). The yellow line in (b) illustrates the deseasonalized trend

228 at Mauna Loa (MLO), same as in (c), in which y-axis is expanded.

229

230   To investigate the contributions of different altitudes to spatial gradients between regions, we divided all

231 measurement data into three layers according to their sampling altitudes: below 2 km, 2 - 5km, and 5 - 8.5 km masl

232 (Fig. 4). Smooth seasonal curves are attained from fitting data with four harmonics using the method described by

233 Thoning et al. (1989). The peak-to-valley amplitudes of the seasonal cycles below 2 km are the largest among the

234 three layers for most regions, with a minimum of 10.3 ppm in SM and a maximum of 25.0 ppm in MW. The

235 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

236 the 5 - 8.5 km layer. We also observe that the seasonal cycle drawdown occurs later in the layers above 2 km (see

237 Fig. S3, which provides similar information as Fig. 4, but seasonal curves from different vertical layers are grouped

238 by regions to facilitate comparisons of the phases of seasonal cycles). The seasonal CO_2 drawdown below 2 km is

239 mainly influenced by terrestrial photosynthesis and gradients are due to local to regional fluxes, with an earlier onset

240 of drawdown in southern regions than in northern regions. The seasonal cycle aloft is damped and lagged compared

241 to the PBL, with influences from throughout the Northern Hemisphere and with spatial gradients likely driven by

242 large-scale transport. The NW, SW, SM, and SE inflow regions have significant delays of more than one month in

243 the 2 - 5 km layer compared with the surface layer, which is likely due to the delayed phase of the seasonal cycle in

244 well-mixed air coming from the oceans. Vertical homogeneity of air over ocean was observed during the HIAPER
245 Pole-to-Pole Observations (HIPPO) aircraft campaign (Wofsy et al., 2011; Frankenberg et al., 2016). As air masses
246 are transported further inland, we observe reduced discrepancies of the timing of CO_2 drawdown between surface
247 and upper layer air (2-5 km), which may be associated with the increased influence of the land surface in the mid-
248 troposphere due to strong convection over land. CO_2 drawdown in the 5 - 8.5 km layers also occurs later than in the
249 2 - 5 km layers in most regions; however, differences between these two layers are small. The declining amplitude
250 and delayed phase of the seasonal cycle with altitude have been noted often (e.g., Tanaka et al., 1983; Ramonet et
251 al., 2002; Gerbig et al., 2003, Sweeney et al. 2015). It demonstrates that there is lot of important information in the
252 vertical profile that is diminished in observations of the total column.

253   We find that the largest horizontal spatial gradients between regions occur below 2 km during summer time
254 (Fig. 4), with a maximum difference of ~15.5 ppm between MW and SM. SM and SW exhibit less pronounced
255 seasonal cycles, which is likely associated with air masses from the Gulf of Mexico and the Pacific Ocean,
256 respectively, whereas MW exhibits a deep summer drawdown (amplitude in seasonal cycles) partially as a result of
257 strong regional forest and crop uptake. Crevoisier et al. (2010) estimated the surface flux over North America using
258 vertical CO_2 measurements and average wind vectors, and reported that annually averaged land carbon fluxes in the
259 western (including SW region) and southern regions (including SM region) were neutral. The SE region also
260 demonstrates a less pronounced seasonal cycle with higher summertime levels compared with other northern
261 regions, which may be due to the sea-breeze influence in summer within PBL. In wintertime, CO_2 levels in NE and
262 MW are higher than in other regions, which result from regional fossil fuel and terrestrial biogenic emissions
263 combined with transport from the west and south.

264   Higher altitude data (above 2 km) exhibit only small spatial gradients. In the 2 - 5 km layer, the largest gradient
265 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
266 shows modeled CO_2 mole fractions from CT2015 for the upper troposphere and above (330 hPa to 0 hPa), which are
267 used to fill in above the aircraft profiles for calculation of total column ΔXCO_2. Spatial gradients in this layer are
268 less than 0.5 ppm, suggesting that the top third of the total column has little contribution to the spatial gradients of
269 the total column.
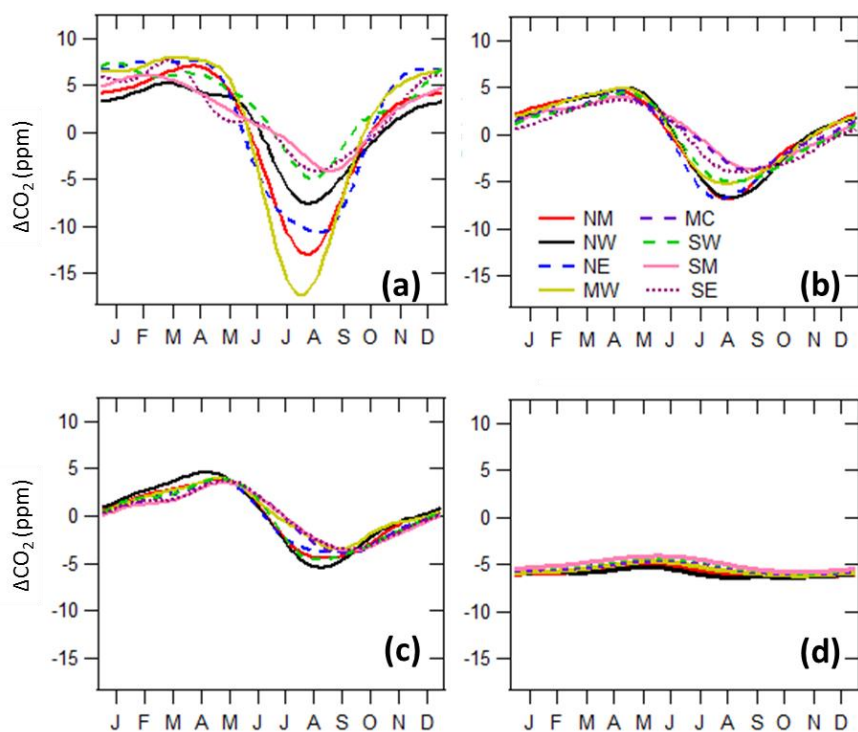
11

Fig. 4. Multi-year (2004-2014) average smooth seasonal curves of 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 (~8.5 km) to 0 hPa (~80 km).

3.2 Long-term mean vertical profiles

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

Figure 5 presents two examples of long term mean profiles with data variability, which is the one standard deviation for each 40 hPa bin of aircraft data or for all flight-day tower data. Variability as large as 20 ppm is seen within the PBL in the MW region in summer, which is due to strong and heterogeneous surface vegetation uptake and ecosystem respiration combined with day-to-day changes in wind direction. All long-term mean monthly vertical profiles are presented in Fig. 6, which shows the mean temporal and vertical variability of CO_2 in each season, and further demonstrates the vertical propagation of seasonal CO_2 due to changes of surface flux. In

289 wintertime, monotonic decrease of CO_2 with altitude can be observed from all regions, in which high PBL CO_2 is

290 mainly driven by surface emissions and reduced vertical mixing (Denning et al., 1998; Stephens et al., 2007).

291 Surface CO_2 decreases dramatically in the growing season in those regions influenced by high plant activity, such as

292 NM and MW regions. For the summer vertical profiles in NE and SE region (east coast of the U.S.), the CO_2 mixing

293 ratio is elevated in the layer under 900 hPa followed by significant decreases in upper layers until 750 hPa, and then

294 increases with altitude until tropopause (Fig. 6). This is likely a result of sea breeze influence. Lower-troposphere air

295 from the sea, lacking terrestrial uptake of CO_2, typically has higher CO_2 in summer compared with inland air.

296 Polluted air previously advected offshore can be brought back along with sea breeze. Without significant vertical

297 mixing over the marine surface, high levels of pollutants remain in those air masses. The convergence of sea breeze

298 with prevailing wind moving offshore may create a period with a stalled frontal structure that can aggregate air

299 pollutants (Banta et al., 2005). The convective internal boundary layer structure of the sea breeze system can

300 significantly reduce mixing height (Miller et al., 2003), and also induces higher CO_2 levels. When the sea breeze is

301 not dominant, air advected from southwest and west (the land) can also bring in polluted air with high CO_2 since this

302 region is downwind of continental U.S. emissions (Miller et al., 2012).
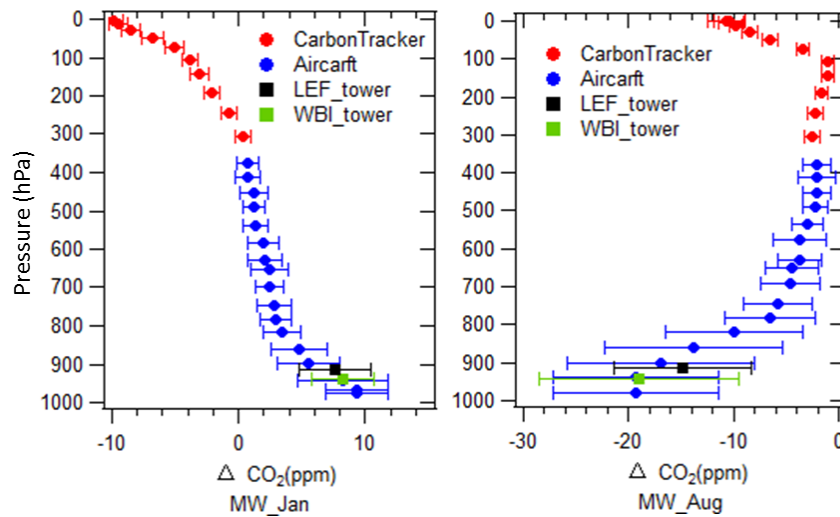

303

304 **Fig. 5**. Long-term mean (2004-2014) average vertical profiles in January (left panel) and August (right panel) in

305 region MW. Error bar shows one standard deviation.
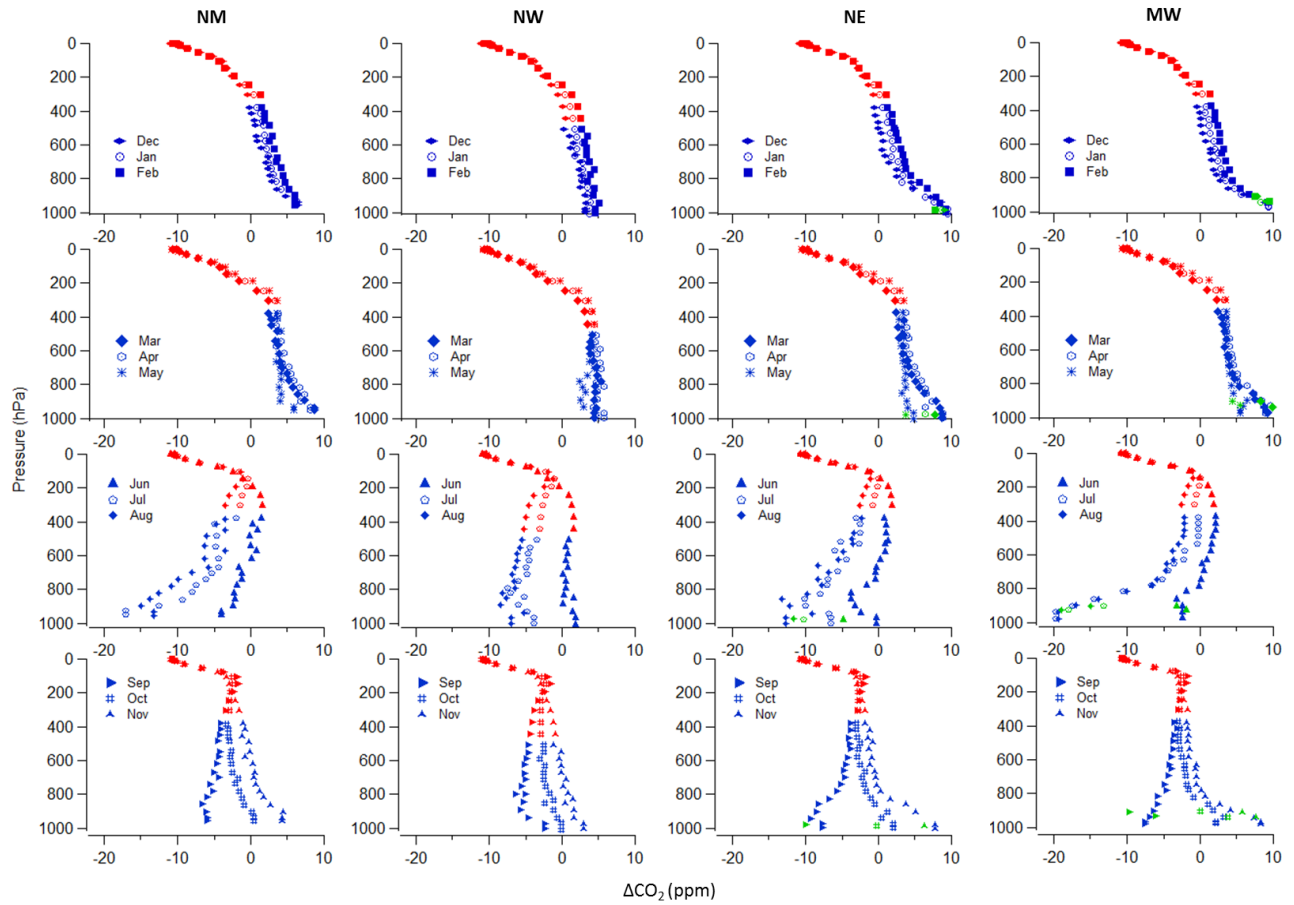
Fig. 6a. Long-term mean (2004-2014) monthly vertical profiles in NM, NW, NE, MW (by column, from left to right). Blue points were calculated from observations, red points were calculated from CT2015, and green points were calculated from tower data.
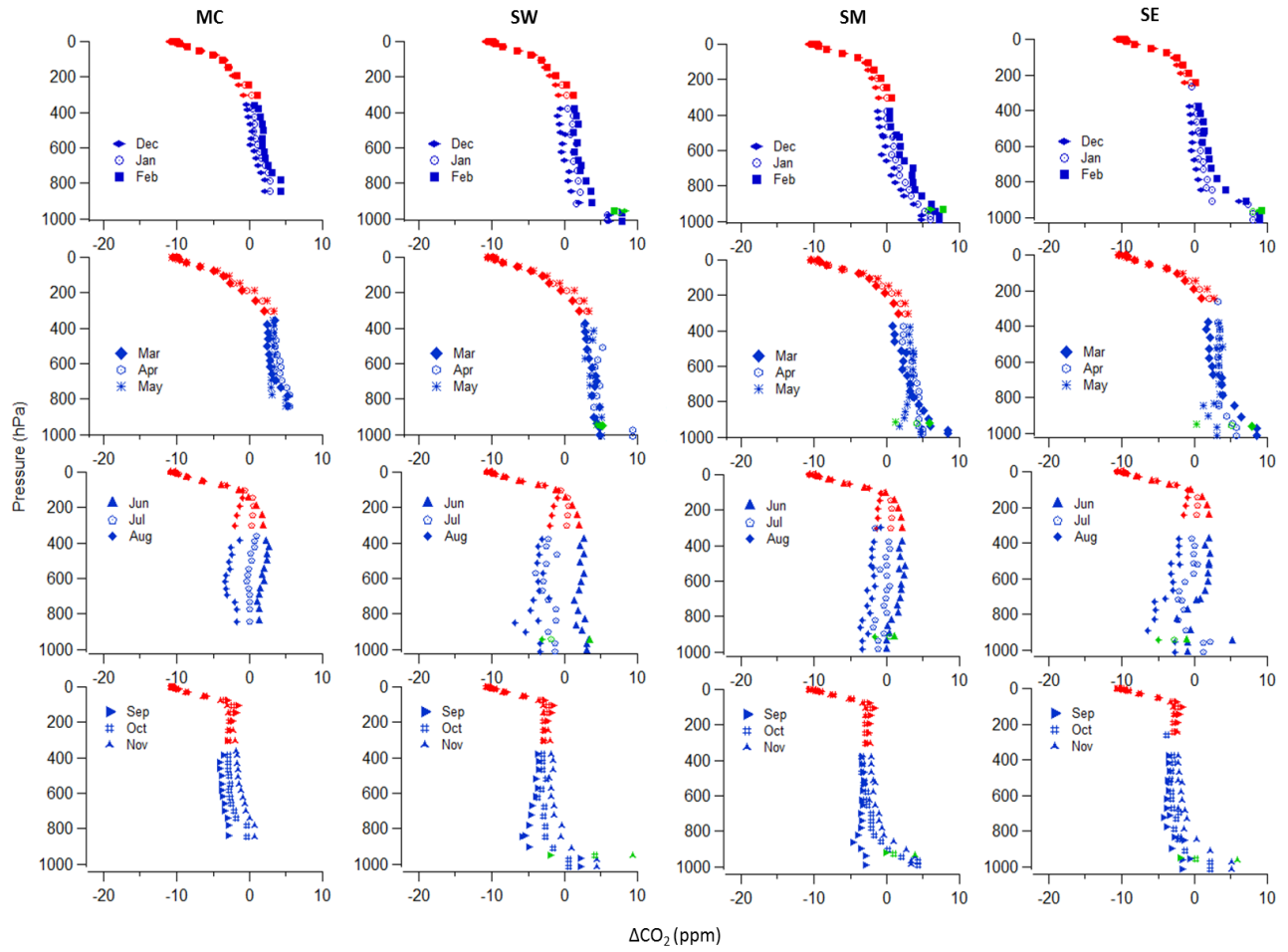
Fig. 6b. Long-term mean (2004-2014) monthly vertical profiles in MC, SW, SM, SE (by column, from left to right). Blue points were calculated from observations, red points were calculated from CT2015, and green points were calculated from tower data.

3.3 Partial column ΔXCO₂ and total column ΔXCO₂

315

316   Seasonal variations of monthly averaged partial column ΔXCO_2 (below 330 hPa) demonstrate maximum values in

317   April and minimum values in August or September (Fig. 7a). The largest amplitude appears in NM, with peak-to-

318   valley difference up to 13.5 ppm. SW, SM, SE, and MC have similar amplitudes of 7-8 ppm, smaller than the other

319   three regions. To evaluate the performance of CT2015 on column ΔXCO_2, CT2015 results are sampled to match the

320   latitude, longitude, altitude and time of actual measurements. Note that aircraft profiles are not assimilated in

321   CT2015, so aircraft data are independent of the CT2015 data assimilation. Figure 7b shows monthly partial columns

322   of ΔXCO_2 calculated from CT2015, which demonstrate good agreement with results from measurements. Only

323   small seasonal biases exist in CT2015, with high bias occurring mostly in spring and early summer and low bias in

324   September and October (Fig. S4). The overall differences of monthly partial column ΔXCO_2 (CT2015 -

325   measurements) mainly fall in the range of -0.64 ppm (5th percentile) to 0.84 ppm (95th percentile) with a mean

326   difference of 0.13 ppm. These differences are of similar magnitude to the uncertainties of partial column ΔXCO_2

327   calculated from the measurements (Fig. S5). It is clear that CT2015 captures the long-term mean variations of both

328   phase and amplitude of partial column XCO_2 reasonably well when compared with well-calibrated measurements

329   across North America.

330

331
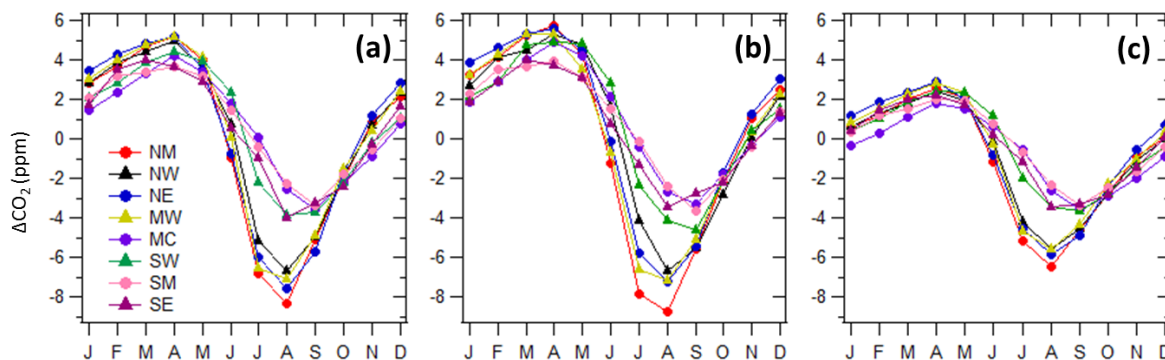
Fig. 7. (a). Partial column ΔXCO_2 calculated from aircraft and tower data; (b). Partial column ΔXCO_2 calculated from CT2015; (c). Total column ΔXCO_2 calculated from aircraft and tower data and including the top layer data from CT2015.

335

336   Total column ΔXCO_2 is presented in Fig. 7c. In regions NW, NM, NE, and MW, seasonal variations of total

337   column ΔXCO_2 are very similar in both phase and amplitude (8-9 ppm peak to valley). For SW, SM, SE, and MC,

338   amplitudes are ~5.5 ppm. The smallest spatial gradients occur during May and October, which result in maximum

339   differences among all regions of only 0.9 and 0.7 ppm, respectively. The largest spatial gradients occur during June,

340   July and August, which result in maximum differences of 2.4, 4.5, and 4.1 ppm, respectively. It is interesting that the

341   deepest seasonal drawdown is seen in region NM, not in region MW that encompasses the very intensive

342 agricultural activities in the U.S. mid-west, which suggest the possibility of strong upwind influence in the NM

343 region. Transported signals have significant influences on total column CO_2. The summer total column ΔXCO_2,

344 represented by the June to August average from CT2015, has a magnitude that is similar to observations with

345 differences no more than 1 ppm (Fig. 8). Based on the seasonal patterns of total column ΔXCO_2 (Fig. 7c) and the

346 summer column ΔXCO_2 (Fig. 8), we can separate the eight regions into two groups. The group with NW, NM, NE,

347 and MW, has ~3 ppm stronger drawdown (larger amplitude) than the group with SW, SM, SE, and MC. For winter

348 total column ΔXCO_2 (December to February average), the maximum spatial difference is only 1.6 ppm, with the

349 highest total column ΔXCO_2 of 1.2 ppm in NE and the lowest value of -0.3 ppm in MC.

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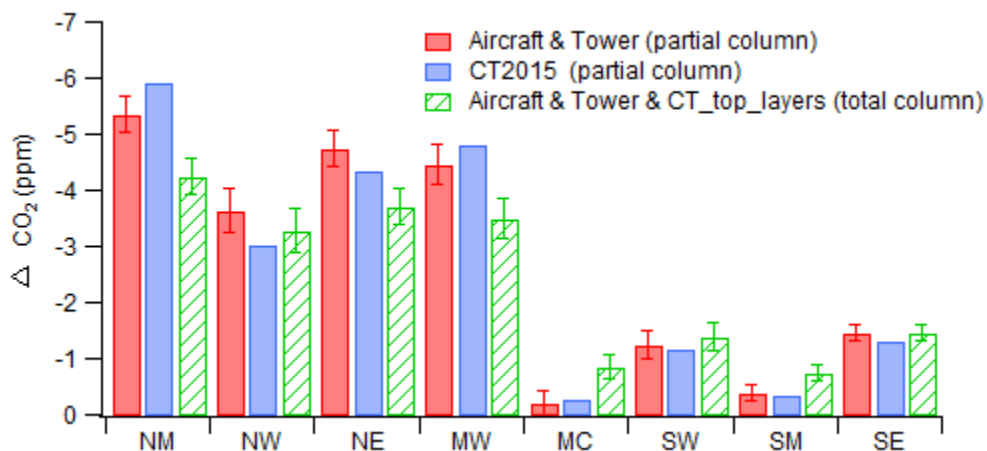
351

352 **Fig. 8**. Long-term mean (2004-2014) June to August partial and total column ΔXCO_2. Error bars represent one

353 standard deviation from the bootstrap uncertainty calculation (see Sect. 2.3).

354

355 **3.4 Influence of large scale circulation**

356 Figure 9 shows long-term mean summer column ΔXCO_2 calculated from CT2015, together with full column

357 ΔXCO_2 from individual aircraft sites. Note that some aircraft sites have less than 11 years of data that CT2015

358 shows in Fig. 9, only aircraft sites with more than 6 years of data are presented, the actual values are provided in

359 Table S2. The fact that total column ΔXCO_2 from CT2015 agrees well with aircraft sites supports the performance

360 of CT2015 on a long-term average basis. The observations show a similar summer spatial pattern, with lower

361 column ΔXCO_2 in the north and northeast regions and higher column ΔXCO_2 in the south and southwest regions

362 (Fig. 9a). Scattered hot spots of high column ΔXCO_2 associated with surface emissions from megacities, or cold

363 spots associated with strong local uptake, are not or just barely visible in the long-term average column ΔXCO_2 map

364 at $1°x1°$ resolution. Instead, the wave-like pattern of column ΔXCO_2 over North America reflects the average large

365 scale circulation. To support our hypothesis on the influence of large scale circulation, we analyze the long term

366 mean wind pattern over North America. We can see that air masses from northwest of the continent bring in low

367 average column ΔXCO_2, while air masses from the south (mainly the subtropical Pacific Ocean and the Gulf of

368 Mexico) bring in high column ΔXCO_2 (Fig. 9b). The zonal gradients over the continent, especially north of 40° N,

369 also reflect long-term average wind patterns; southwest wind corresponds to higher column ΔXCO_2 over the western

370 part of the continent until the wind direction shifts to west-northwest over the eastern part of the continent. This

371 wind pattern matches well with the geographic division of the over/under -3 ppm areas colored in green/blue in the

372 column ΔXCO_2 map (Fig. 9b). Figure 9c and 9d shows partial column averages for free troposphere (800-330 hPa)

373 and lower troposphere (below 800 hPa), respectively. The free troposphere spatial gradient also demonstrates a

374 wave-like pattern. A previous study on the total column CO_2 from ground based Total Carbon Column Observation

375 Network (TCCON) found strong correlation between the mid-latitude column CO_2 and synoptic-scale variation of

376 potential temperature (θ, at 700 hPa), a dynamic tracer for adiabatic air transport (Keppel-Aleks et al., 2012). Thus

377 they also propose that the variations in column CO_2 are mainly driven by large-scale flux and transport. Analysis of

378 the interannual variability of the seasonal cycle amplitudes of column CO_2 in North Hemisphere has also found

379 significant contribution of large-scale circulations to the north-south gradient (Wunch et al., 2013).

380   The strong drawdown over northeast North America in summer is a consequence of long-range transport of low

381 CO_2 from northeast Eurasia, in addition to regional terrestrial uptake.   Sweeney et al. (2015) notes well-mixed

382 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°,

383 -147.29°) sites and suggests that air coming across the Pacific was strongly influenced by Asian surface fluxes

384 before being vertically homogenized as it passed over the Pacific Ocean. This well-mixed air forms an important

385 boundary condition in the column CO_2 of air coming into the North American continent. This was best illustrated at

386 sites like PFA where the summertime minimum in CO_2 significantly preceded maximum ecosystem uptake of CO_2,

387 implying significant influence of transported air from lower latitude regions from Asia.   We further conduct an

388 experiment using Carbon Tracker to investigate the importance of this effect. A control run and a "masked run" are

389 conducted for 2010-2012, in which the Eurasian boreal flux is turned on/off. The MLO CO_2 trend from each model

390 scenario is used as reference background and thus removed before total column ΔXCO_2 calculation. Figure 10 shows

391 the results for 2012 summer, which is an average summer when compared with the 2004-2014 mean pattern (Fig. 9

392 and Fig. 11). The maximum north-south difference reduces to ~2.5 ppm after we turn off the Eurasian boreal flux,

393 compared with ~5 ppm from the control run. In both control and masked scenarios, the free troposphere partial

394 ΔXCO_2 demonstrates similar spatial patterns as for total column ΔXCO_2 (Fig. S6). This result combined with results

395 from Sweeney et al. (2015) demonstrates that the transport of low CO_2 resulting from large summertime Eurasian

396 boreal uptake has a large contribution on the overall summer total column CO_2 decrease in North America.

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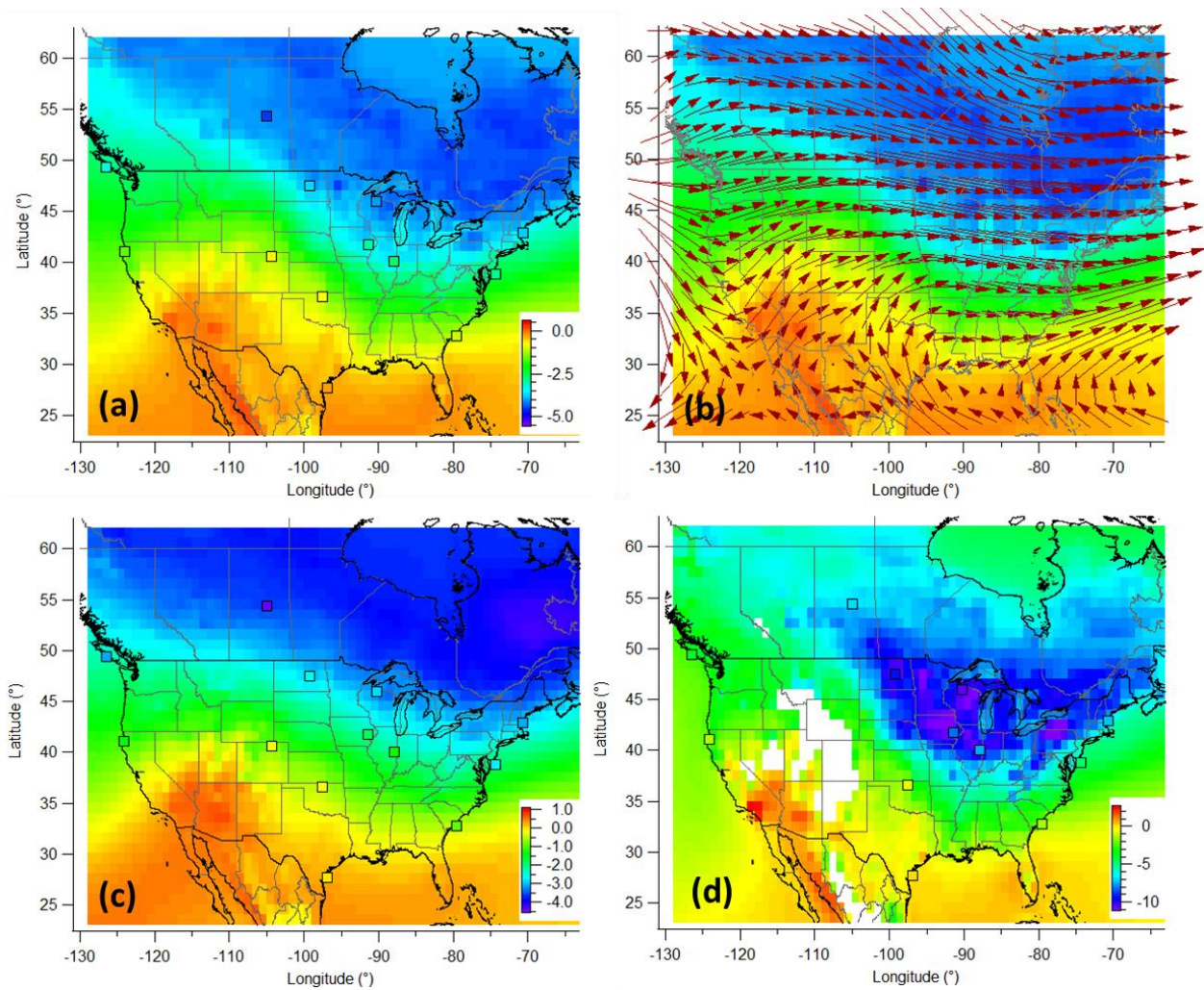
Fig. 9. Long-term mean (2004-2014) June-August total column ΔXCO_2 from CT2015 in $1° \times 1°$ spatial resolution with total column ΔXCO_2 for 13 individual aircraft sites in squares (a), and CT2015 column ΔXCO_2 overlaid with pressure-weighted (1000 hPa to 500 hPa) mean wind vectors for the same period (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.
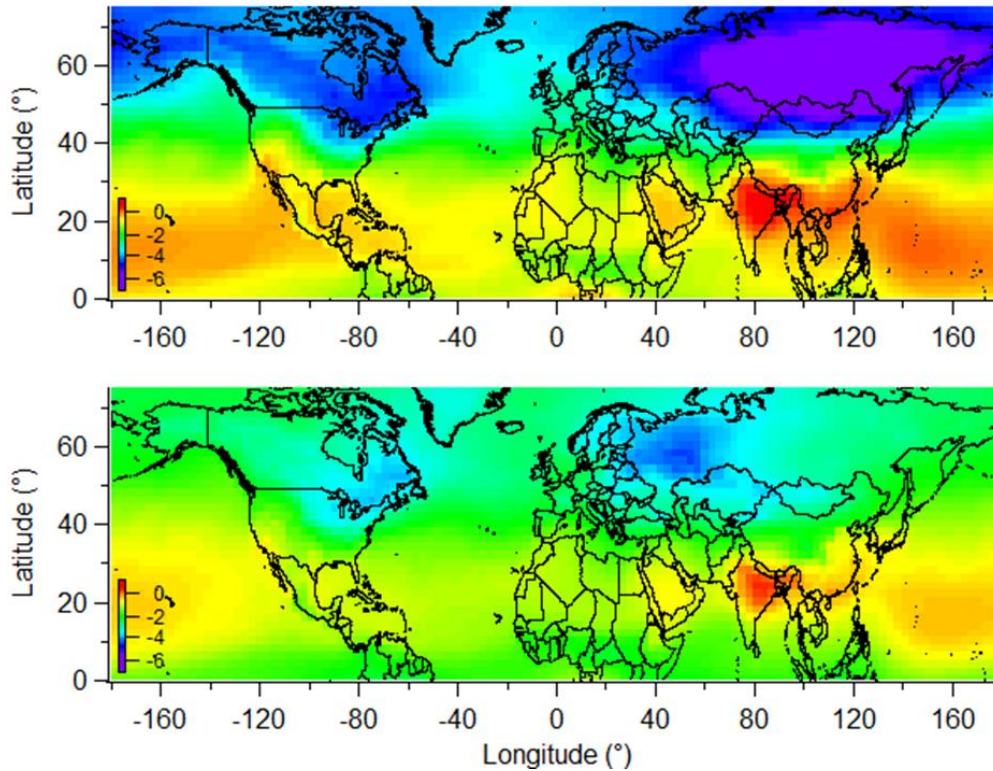
Fig. 10. Total column ΔXCO_2 from Carbon Tracker control (top panel) and masked (bottom panel, Eurasian boreal flux is masked) runs for 2012 June-August ($3° \times 2°$ spatial resolution). MLO trend from each individual scenario is removed before the ΔXCO_2 calculation. Same color scale is used as in Fig. 9a. Partial column ΔXCO_2 patterns for free troposphere (800 to 330 hPa) and lower troposphere (below 800 hPa) are provided in SI.

**3.5 A comparison with apparent gradients over Europe**

Figure 11 shows the climatological June - August mean modeled global column ΔXCO_2 map in $3° \times 2°$ 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 ΔXCO_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 ΔXCO_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

20

models produced similar fluxes over the European region (see both websites for detailed fluxes). The $3^o \times 2^o$ grid from CT2015 is not likely responsible for a much smoother pattern for Carbon Tracker, compared with the $2^o \times 2^o$ grid from satellite retrievals (Reuter et al., 2014) . The North America region on the $3^0 \times 2^0$ grid in Fig. 11 shows similar pattern as the $1^o \times 1^o$ 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/~~date~~ hour (within 1 hour) as ~~by~~ in SCIAMACHY BESD (v02.00.08) data (Reuter et al., 2011). The 8~~3~~-year ~~(2005-2007)~~ mean pattern shows much smaller gradients (3-4 ppm maximum) without significant hot/cold spots at those locations as SCIAMACHY (Fig. S7). Sampling bias is unlikely the main reason for the unphysical column XCO_2 spatial pattern from SCIAMACHY. Since the satellite retrievals in Reuter et al. (2014) appear to show unrealistic column XCO_2 spatial gradients over Europe, they should not be used to derive any estimates of a European carbon sink. 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, ~~which shows 0.03 ± 2.33 Pg C yr^{-1} for Europe,~~ should be more trustworthy than fluxes estimated based on unrealistic spatial pattern. However, the European carbon sink is still inconclusive; the discrepancies among different methods and results are further discussed by Reuter et al. (2017). Increasing the amount of highly precise observations such as the well-calibrated surface measurements and vertical profiles can greatly help to estimate the carbon sink.
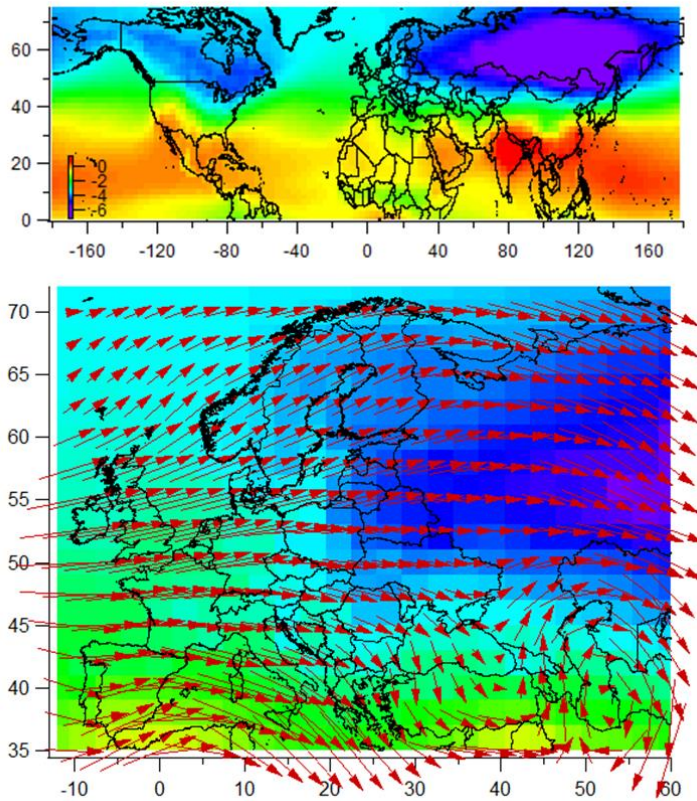
Fig. 11. Long-term mean (2004-2014) June - August total column ΔXCO_2 from CT2015 (top panel) in $3° \times 2°$ spatial resolution, and detail for Europe overlaid with pressure-weighted (1000 hPa to 500 hPa) mean wind vectors for the same period (bottom panel). The color scale is the same as in Fig. 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).

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_2 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_2 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_2 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_2 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 ΔXCO_2 was calculated from the long-term mean vertical profiles. By comparing the partial column

465 ΔXCO_2 from measurements with those from CT2015, we verify that CT2015 captures the long-term mean patterns
466 of both phase and amplitude of partial ΔXCO_2.

467   Large spatial gradients of ΔXCO_2 only appeared in summer, during which time the north and northeast regions
468 had ~3 ppm stronger drawdowns than the south and southwest regions. Scattered hot spots of high column ΔXCO_2
469 associated with surface emissions from megacities, or cold spots associated with strong local uptake, are not or just
470 barely visible in the long-term average column ΔXCO_2. Instead, the wave-like pattern of column ΔXCO_2 over North
471 America matches well with the average large scale circulation. A CarbonTracker experiment to investigate the
472 impact of Eurasian long-range transport suggests that the large summer time Eurasian boreal flux alone contributes
473 about half of the north-south column ΔXCO_2 gradient across North America. Considering the transported signals
474 from other upwind regions, including northern Canada, we expect that the transported signals have the overall
475 largest contribution to the total column ΔXCO_2 spatial gradient.

476 **Author contributions**

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

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