

1 **Estimating regional greenhouse gas fluxes: An uncertainty analysis of**
2 **planetary boundary layer techniques and bottom-up inventories**

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14

15 **Abstract**

16 Quantification of regional greenhouse gas (GHG) fluxes is essential for establishing mitigation
17 strategies and evaluating their effectiveness. Here, we used multiple top-down approaches and
18 multiple trace gas observations at a tall tower to estimate regional-scale GHG fluxes and evaluate
19 the GHG fluxes derived from bottom-up approaches. We first applied the eddy covariance,
20 equilibrium, inverse modeling (CarbonTracker), and flux aggregation methods using three years
21 of carbon dioxide (CO₂) measurements on a 244-meter tall tower in the Upper Midwest, USA.
22 We then applied the equilibrium method for estimating CH₄ and N₂O fluxes with one-month
23 high-frequency CH₄ and N₂O gradient measurements on the tall tower and one-year
24 concentration measurements on a nearby tall tower, and evaluated the uncertainties of this
25 application. The results indicate that: 1) The flux aggregation, eddy covariance, the equilibrium
26 method, and the CarbonTracker product all gave similar seasonal patterns of the regional CO₂
27 flux (10^5 - 10^6 km²), but that the equilibrium method underestimated the July CO₂ flux by 52-69%.
28 2) The annual budget varied among these methods from -54 to -131 g C-CO₂ m⁻² yr⁻¹, indicating
29 a large uncertainty in the annual CO₂ flux estimation. 3) The regional CH₄ and N₂O emissions
30 according to a top-down method were at least six and two times higher than the emissions from a
31 bottom-up inventory (Emission Database for Global Atmospheric Research), respectively. 4) The
32 global warming potentials of the CH₄ and N₂O emissions were equal in magnitude to the cooling
33 benefit of the regional CO₂ uptake. The regional GHG budget, including both biological and
34 anthropogenic origins, is estimated at 7 ± 160 g CO₂ eq m⁻² yr⁻¹.

35 **Keywords:** eddy covariance; equilibrium method; greenhouse gas; inverse method; land surface
36 flux; tall tower

37 **1. Introduction**

38 Although quantifying GHG fluxes at the regional scale (10^2 - 10^6 km²) is essential for
39 coordinating GHG mitigation strategies, observations and flux information at these relevant
40 scales are still extremely limited (e.g., Chen et al., 2008; Nisbet and Weiss, 2010). To fill this
41 scale gap, some researchers build ecosystem models and aggregate the modeled flux according to
42 land information (e.g., Desai et al., 2008; Tang et al., 2012; Xiao et al., 2008), while others use
43 GHG concentration observations in combination with atmospheric transport models to derive the
44 land surface flux (Lauvaux et al., 2012; Peters et al., 2007). The aggregation method is a bottom-
45 up approach. Another bottom-up method is the IPCC national GHG inventory system (IPCC,
46 2006) based on emission factors and data concerning anthropogenic activities. The bottom-up
47 applications are relatively easy to implement; however, they require independent verification,
48 because uncertainties in land cover, anthropogenic activity, vegetation flux, and emission factors
49 can lead to large biases (Chen et al., 2008; Levy et al., 1999). Hence, there is a strong motivation
50 for using top-down methods to provide an independent constraint on the regional fluxes.

51 There are several top-down methods for estimating regional GHG fluxes, including tall-tower
52 eddy covariance (Davis et al., 2003), the equilibrium boundary layer approach (Bakwin et al.,
53 2004; Betts et al., 2004; Desai et al., 2010; Helliker et al., 2004), and inverse modeling (Peters et
54 al., 2007). Each method uses different assumptions, has inherent advantages and disadvantages,
55 and is sensitive to different parameters. Eddy covariance (EC) provides a direct measurement of
56 the flux using measurement of the wind fluctuations and the scalar of interest. EC has been used
57 for CO₂ flux measurement on tall towers (Davis et al., 2003; Haszpra et al., 2005), while few
58 tall-tower flux observations of CH₄ and N₂O have been carried out due to instrument limitations
59 (Desai et al., 2012) and the relatively large uncertainty for these measurements (20–300% for

60 CH₄, 30–1800% for N₂O) (Kroon et al., 2010). Based on the mass balance in the atmospheric
61 boundary layer, the equilibrium method assumes that the exchange at the top of the boundary
62 layer and the exchange at the land surface are in equilibrium over periods longer than about 1
63 month (Betts, 2000). The largest source of uncertainty of this method lies in determining the
64 background concentration above the boundary layer and the entrainment rate at the top of the
65 boundary layer. Inverse modeling determines the land surface flux using atmospheric transport
66 models that are constrained by observed trace gas concentrations. The prior land surface flux,
67 land surface observations, the meteorological inputs, and atmospheric transportation schemes are
68 all important for determining the accuracy of the modeled flux (Peters et al., 2007). As a result,
69 the deficiency in any of these four factors can limit the accuracy of the model.

70 In this study, we used several top-down approaches to evaluate the bottom-up fluxes of CO₂,
71 CH₄, and N₂O for a region dominated by agriculture. The inter-comparison of multiple
72 techniques was used to identify systematic biases of each method and constrain the overall
73 uncertainties. We first used CO₂ to evaluate the equilibrium boundary layer method against tall-
74 tower eddy covariance, flux aggregation, and the flux produced by an inverse model. We then
75 applied the equilibrium method to estimate the CH₄ and N₂O fluxes. The final task was to
76 compare the CH₄ and N₂O fluxes with two bottom-up emission inventories: 1) EDGAR42
77 (European Commission, Joint Research Centre [JRC]/Netherlands Environmental Assessment
78 Agency [PBL], Emission Database for Global Atmospheric Research [EDGAR], release version
79 4.2, <http://edgar.jrc.ec.europa.eu>, 2011), an inventory dataset used widely in atmospheric models
80 (Jeong et al., 2012); and 2) a national GHG inventory developed by the U.S. Environmental
81 Protection Agency (U.S. EPA, 2014).

82

83 **2. Data and Methods**

84 **2.1 Research site**

85 The boundary layer observations were made on a 244 m communication tower (KCMP tower)
86 located at the Rosemount Research and Outreach Center, University of Minnesota, about 25 km
87 south of Minneapolis/Saint Paul (44°41'19"N, 93°4'22"W). According to the U.S. Department of
88 Agriculture Crop Data Layer data in 2009, the landscape around the tall tower was dominated by
89 cropland, which accounted for 41% of the land cover within the 10 km radius of the tower and
90 37% within the 600 km radius. Corn and soybean were the dominant crop species, accounting for
91 55% and 38% of the cropland, respectively. About 40% of the land within the 600 km radius was
92 covered by forest, grassland and pasture. The other land use was comprised of developed land,
93 wetland, and open water. The land cover pattern described here for 2009 had a smaller corn to
94 soybean ratio than that reported by Griffis et al. (2010) for 2007. This difference was mainly
95 attributed to more corn plantation in 2007 stimulated by increased ethanol biofuel demand.

96 **2.2 Mixing ratio data**

97 CO₂ mixing ratios at the 32 m, 56 m, 100 m, and 200 m heights above the ground were measured
98 by a tunable diode laser analyzer (TDL) (model TGA 100A, Campbell Scientific Inc., Logan,
99 UT, USA) (Griffis et al., 2010). Air at these levels was drawn down by a pump (model DOA-
100 V502A-FB, Gast Group Inc., Benton Harbor, MI, USA) through four Synflex tubes (6.25 mm ID)
101 at a line pressure of 60 kPa and at a flow rate of 16 L min⁻¹. The air was sampled sequentially,
102 each for 30 s. The sampled air was dried prior to analysis using a Nafion drier and brought to a
103 common temperature. The CO₂ measurement was calibrated for every measurement cycle
104 against the National Oceanic and Atmospheric Administration-Earth System Research

105 Laboratory (NOAA-ESRL) standards. The hourly precision of the CO₂ measurement was
106 approximately 0.03 ppm.

107 In addition, an intensive campaign was carried out from August 30 to September 25 (DOY 243 -
108 269), 2009. During this campaign, we measured CO₂, H₂O, CH₄, and N₂O mixing ratios at the
109 200 m and 3 m height on the tower. Air was drawn from these heights at a flow rate of 1.3 L min⁻¹
110 and 0.9 L min⁻¹, respectively, through two Synflex tubes (6.25 mm ID). A portion (0.6 L min⁻¹)
111 of this flow was delivered to an infrared gas analyzer (IRGA model LI-6262, LI-COR, Lincoln,
112 NE, USA) for CO₂ and H₂O mixing ratio measurements, and a small amount (180 mL min⁻¹) was
113 delivered to another TDL for CH₄ and N₂O measurements. Measurement precisions for CO₂,
114 CH₄, and N₂O were 0.2 ppm, 1.2 ppb, and 0.5 ppb, respectively. The IRGA was manually
115 calibrated with a standard CO₂ gas (391.03 ± 0.03 ppm) and a dew point generator (LI-610, LI-
116 COR) at the beginning of the experiment. The accuracy of its measurement was improved in
117 post-field analysis by adding offsets so that its 200-m reading matched that registered by the
118 TDL CO₂ analyzer for the same height. The TDL for N₂O and CH₄ measurement was plumbed
119 to a four-port manifold that used a switching sequence in the order of 200 m, 3 m, calibration
120 zero and calibration span, with 30 s spent on each port and the first 15 s after each switching
121 omitted from the analysis. The N₂O concentration of the calibration span was traceable to a
122 NOAA-ESRL gold standard. The CH₄ concentration of the calibration span was calibrated
123 against a known standard provided by a local supplier and was also traceable to the NOAA-
124 ESRL standard scale.

125 **2.3 Eddy covariance data**

126 A closed-path EC system installed at the height of 100 m on the tower was used to measure the
127 CO₂ flux from 2007 to 2009 (Griffis et al., 2010). This system consisted of a 3-D sonic

128 anemometer-thermometer (model CSAT3, Campbell Scientific Inc.) and the TDL analyzer
129 (model TGA 100A, Campbell Scientific) for CO₂ concentration. The sample tubing was 125 m
130 long (6.25 mm ID, Synflex), which resulted in a typical lag time of 11 s, with Reynolds numbers
131 exceeding 3500. Fluctuations in the velocities and concentrations were recorded at 10 Hz, and a
132 block averaging time of 60-min was used to capture the dominant flux-containing frequencies.

133 Further, in 2009 two closed-path eddy covariance systems were used at two 10-m towers in the
134 middle of corn (G21) and soybean fields (G19) (Baker and Griffis, 2005) about 3 km away from
135 the tall tower. They recorded half-hourly fluxes of CO₂ and H₂O.

136 **2.4 Top-down flux estimation methods**

137 **2.4.1 Tall-tower eddy covariance**

138 Briefly, the tall-tower CO₂ flux was determined as the sum of the eddy covariance term
139 measured at the 100-m height ($\overline{w'c'}$) and the storage term between the land surface and this
140 height (F_S).

$$141 \quad F_{EC} = \overline{w'c'} + F_S \quad (1)$$

142 Here, we assume that horizontal and vertical advection were negligible (Davis et al., 2003;
143 Griffis et al., 2010). Wind statistics and fluxes were transformed into the planar fit coordinate
144 system (Lee et al., 2004). Eddy fluxes were computed using the maximum covariance method
145 with strict limits on window size based on manifold pressure and flow rates. Flux losses
146 attributed to a combination of sensor separation, sonic path averaging, tube attenuation, and
147 block averaging were estimated using the analytical model of Massman (2000). These losses
148 typically ranged between 5% and 20%. A detailed description of the eddy covariance system and
149 flux calculation can be found in Griffis et al. (2010).

150 The eddy covariance method does not perform well in stable atmospheric conditions and friction
151 velocity (u^*) is commonly used as a quality control for such conditions (Davis et al., 2003;
152 Goulden et al., 1996). In this study, we discarded the nighttime flux data when u^* was less than
153 0.10 m s^{-1} , which is a threshold often used for agricultural environments (Baker and Griffis, 2005;
154 Griffis et al., 2005).

155 Large negative fluxes in the early morning have been observed at many eddy covariance tower
156 sites, and it may lead to an overestimation of CO_2 uptake during the growing season by as much
157 as 20% (Anthoni et al., 1999; Davis et al., 2003; Yi et al., 2000). Davis et al. (2003) suggested
158 that this bias is caused by horizontal and vertical advection, and it can be corrected by excluding
159 the negative CO_2 flux that exceeds a pre-defined level. In this study, we excluded the morning
160 (06:00 and 10:00 LST) data when the storage term was large (i.e. $F_S < -4 \mu\text{mol m}^{-2} \text{ s}^{-1}$). This
161 storage term screening reduced the estimated CO_2 uptake during the growing season (May to
162 September) by 18% and is consistent with that reported in the literature (Davis et al., 2003; Yi et
163 al., 2000). Details about the calculation of the storage term and a discussion about the data-
164 screening standard for the negative storage term in early morning is reported in the
165 supplementary materials (Section S1).

166 The monthly CO_2 flux was determined by the mean of the composite diurnal variation of the CO_2
167 flux. In 2009, excluding the malfunctioning of the instrumentation, the available data was 78%.
168 The u^* and storage term screening eliminated an additional 12% and 2% of the data, respectively.
169 We estimated the monthly mean from the diurnal composite of the CO_2 flux based on valid
170 observations (Section S2 in supplementary materials).

171 2.4.2 Equilibrium method

172 The equilibrium method (EQ) provides a way to quantify regional trace gas fluxes from mixing
173 ratio measurements in the boundary layer (Bakwin et al., 2004; Betts, 2000; Betts et al., 2004;
174 Denmead et al., 1996; Desai, 2010; Helliker et al., 2004; Williams et al., 2011). So far, this
175 method has been applied to CO₂ and N₂O (Griffis et al., 2013) but not to CH₄. The EQ method
176 assumes that, over relatively long timescales (weeks), the diurnal dynamics of boundary layer
177 processes can be ignored and the boundary layer reaches statistical equilibrium (Griffis et al.,
178 2013; Helliker et al., 2004). Therefore, the averaged horizontal advection and storage are
179 negligible in the boundary layer budget (Williams et al., 2011) and the land surface flux (F_{Eq}) is
180 in balance with the exchange at the top of the boundary layer, as

$$181 \quad F_{Eq} = \rho W(c_+ - c_m) \quad (2)$$

182 where c_+ and c_m are the mixing ratio of CO₂, CH₄ or N₂O above and within the boundary layer,
183 respectively and ρ and W are air density and the vertical velocity, respectively, at the top of
184 boundary layer. Here c_+ was assumed as the concentration measured at Niwot Ridge (NWR,
185 40°3'11"N, 105°35'10"W) CO, USA, which is the closest background site operated by NOAA
186 (Conway et al., 1994) and is upwind of KCMP tower in the Ferrel cell. c_m was the concentration
187 at 200 m measured by TDL analyzers and calibrated to the NOAA-ESRL standards. The
188 concentrations used in the calculation were the composite diurnal variations for each month in
189 the case of CO₂ and the diurnal composites for the intensive campaign in the case of CH₄ and
190 N₂O. The equilibrium method was used for calculating the CO₂ flux from 2007 to 2009.
191 However, due to availability of data, the comparison among methods is limited to the 2009 CO₂
192 fluxes.

193 We used the following two methods to determine ρW (Helliker et al., 2004) for the three GHGs:

$$\rho W = \frac{F_w}{c_{w,+} - c_{w,m}} \quad (3)$$

194

$$\rho W = -\frac{\Omega}{g \cdot M_{air}} \quad (4)$$

195 where F_w and $c_{w,m}$ are the water vapor flux and mixing ratio measured at 100 m and 200 m on
196 the tall tower, respectively, $c_{w,+}$ and Ω are the water vapor mixing ratio and the pressure vertical
197 velocity (in units of Pa s^{-1}) at the 700 hPa level in the NCEP/NCAR Reanalysis-2 data, g is
198 gravitational acceleration, and M_{air} is molecular mass of dry air (g mol^{-1}). ρW calculated from
199 equation 3 is essentially the same as equation 4 under the EQ assumptions, because large scale
200 synoptic subsidence dominates the exchange at the top of the boundary layer (Helliker et al.,
201 2004). In addition, for CH_4 and N_2O , we also used CO_2 as a tracer to determine ρW :

$$\rho W = \frac{F_C}{c_{C,+} - c_{C,m}} \quad (5)$$

202 where F_C is the CO_2 flux measured by the EC system on the tall tower, $c_{C,+}$ and $c_{C,m}$ are the CO_2
203 mixing ratio measured at the NWR background site and at the 200 m level on the tall tower,
204 respectively.

205 **2.4.3 Inverse modeling**

206 We used the CO_2 flux product from the global inversion model CarbonTracker 2011_oi (CT)
207 (Peters et al., 2007 with updates documented at <http://carbontracker.noaa.gov>) as a reference to
208 compare with the flux determined with the methods described above. This product provides
209 three-hourly CO_2 fluxes from 2000-2010 at a spatial resolution of 1° by 1° , so the number of grid

210 points within the 100 km, 200 km, 300 km, and 600 km radii of the tall tower is 2, 10, 25, and 90,
211 respectively.

212 The inversion CO₂ flux consists of fossil fuel burning, fire, land, and ocean flux. The CO₂ flux
213 from fossil fuel was the average of two fossil fuel CO₂ emission datasets: one is the legacy
214 CarbonTracker fossil fuel product using the global total from the Carbon Dioxide Information
215 and Analysis Center (CDIAC, Boden et al. 2011) and the spatial distribution from EDGAR; and
216 the other is the ODIAC (Open-source Data Inventory for Anthropogenic CO₂) emission product
217 reported by Oda and Maksyutov (2011).

218 **2.5 Flux aggregation (FA)**

219 The regional trace gas flux (F_{FA}) can be estimated by aggregating sectorial and spatial fluxes
220 using sectorial statistics and land cover information (Chen et al., 2008; Desai et al., 2008; Nisbet
221 and Weiss, 2010; Tang et al., 2012). The total CO₂ flux from the landscape is the sum of the
222 anthropogenic flux and biological flux. In this study, the anthropogenic flux (F_{ant}) was the
223 prescribed fossil fuel flux in the CarbonTracker product for a target region. The biological flux
224 was calculated by aggregating the CO₂ flux from six major land cover types.

$$225 \quad F_{FA} = F_{ant} + \sum_{i=1}^6 \text{frac}_i \cdot F_{bio,i} \quad (6)$$

226 In this equation, frac_i is the fraction of land cover type i for a target region, and $F_{bio,i}$ is the CO₂
227 flux from land cover type i . The six land cover types are cropland (corn and soybean), forest,
228 grassland/pasture, wetland, open water, and developed land.

229 In order to compare with the fluxes from top-down methods, we estimated the bottom-up flux
230 using the flux aggregation method within the tall tower footprint. Various methods have been
231 developed for determining the footprint of the concentration or eddy flux measurement (Chen et

232 al., 2009; Kljun et al., 2004; Lin et al., 2003; Vesala et al., 2008). We used two footprint methods.
233 The first method is based on determining an equally-weighted circular footprint centered at the
234 tall tower. This method assumes that the area within each circular footprint has the same
235 influence on the flux measured at the tall tower despite its distance from the tower, and therefore,
236 $frac_i$ is the fraction of land cover type i within a certain radius around the tall tower. We tested
237 the radii from 5 km to 600 km because the fetch of the EC flux footprint is thought to be 10
238 (during strong convection) to 100 (during neutral or stable condition) times the measurement
239 height (Horst and Weil, 1992; Davis et al., 2003), however, some studies also suggest that the
240 fetch-to-measurement-height ratio is much higher than 100 when the flux is measured at a high
241 level (e.g. higher than 20 m) (Gash, 1986; Leclerc and Thurtell, 1990). The second footprint
242 method we applied derives the footprint from the Stochastic Time-Inverted Lagrangian Transport
243 model (STILT, Lin et al., 2003). During September, when the intensive campaign was carried
244 out, we released 100 air parcels hourly at the tower and transported these parcels backward for
245 two days. The distribution of the air parcels determined the tall tower footprint. (This footprint
246 represents the source area of the EQ flux calculated based on the 200 m concentration.)
247 Therefore, $frac_i$ was determined by overlaying the weighted footprint map with the land cover
248 map. The values of $frac_i$ from these two different methods are summarized in Table S3 of the
249 supplementary materials.

250 The cropland CO₂ flux was the weighted average of the flux measured with EC in a soybean
251 field and a cornfield near the tall tower as described above. The forest CO₂ flux was obtained
252 from the AmeriFlux data archive (Level 2 data) for the deciduous forest site at the University of
253 Michigan Biological Station (UMBS), 662 km northeast of the tall tower (Curtis et al., 2005;
254 Schmid et al., 2003). The grassland CO₂ flux was also from USIB2 AmeriFlux for Fermi Prairie,

255 Illinois, 503 km southeast of the tower (Gomez-Casanovas et al., 2012). Each of the three land
256 cover types was measured by EC flux towers in 2009, and showed different seasonal patterns of
257 CO₂ flux (Figure 1). The biological CO₂ flux from wetland, open water, and developed land was
258 considered as negligible in this study, because these three land cover types only accounted for
259 about 20% of the tall tower footprint, and the reported annual CO₂ fluxes from those land cover
260 types were not significantly different from zero or relatively small (Striegl et al., 1998; Knoll et
261 al., 2013; Olson et al., 2013). For example, Olson et al. (2013) reported that a temperate peat
262 land in northern Minnesota, USA was a small net sink of CO₂ in 2009 (-26.8 ± 18.7 g C-CO₂ m⁻²
263 yr⁻¹), only about 5% of the CO₂ flux from a cornfield, and about 10% of the biogenic flux in the
264 tall tower footprint. Considering the land fraction of peat land in the tall tower footprint, the CO₂
265 flux was estimated to be less than 1% of the biogenic flux.

266 **3. Results**

267 **3.1 Constraints on the regional CO₂ flux**

268 The tall tower EC CO₂ flux exhibits a strong seasonal pattern (Figure 2). From October to April,
269 the landscape was a net source of CO₂, and the averaged efflux was 0.68 ± 0.10 μmol m⁻² s⁻¹ (the
270 mean and standard deviation of the three annual values from 2007 to 2009). From May to
271 September, the landscape was a sink of CO₂, reaching a peak uptake in July at the rate of $-3.68 \pm$
272 0.99 μmol m⁻² s⁻¹. There was no monthly mean data for June 2007 and June 2009 due to
273 measurement problems. Since June is the only month that has missing CO₂ flux data for 2009,
274 we gap-filled it according to the flux values observed in May to July 2008 and 2009. The annual
275 cumulative flux in 2008 and 2009 was -24 and -131 g C-CO₂ m⁻² yr⁻¹, respectively.

276 In order to test the size of the region that our monthly averaged EC flux represents, we first used
277 CT and FA methods to estimate the CO₂ flux with the equally-weighted circular footprint for a
278 radius up to 600 km and compared them with the EC flux. The monthly flux values from these
279 methods for the range of radii correlated well with the EC flux ($r > 0.9$, $p < 0.001$) (Table 1),
280 suggesting the land surface flux was relatively homogeneous and was dominated by the seasonal
281 pattern of the biological flux. Further, to test the accuracy of the estimation, the Nash-Sutcliffe
282 efficiency (NSE) was calculated (Nash and Sutcliffe, 1970), as

$$\text{NSE} = 1 - \frac{\sum(o_i - m_i)^2}{\sum(o_i - \bar{o})^2} \quad (7)$$

283 where o_i is the EC flux and m_i is the regional mean flux from CT or FA. The summation is
284 performed over all months and the overbar denotes the mean over these months. It is considered
285 a very good fit when $\text{NSE} > 0.75$, and a good fit when $0.65 < \text{NSE} \leq 0.75$ (Moriassi et al., 2007).
286 The results show that the EC flux agrees very well with the regional mean flux from both the CT
287 and FA methods with a 200 km radius or larger ($\text{NSE} > 0.80$). Furthermore as the radius
288 increased from 200 km to 600 km, the CT and FA fluxes did not change significantly. The CT
289 and FA fluxes within a 100 km radius were more positive than the EC flux, mainly due to the
290 strong local anthropogenic emission from the Minneapolis/Saint Paul urban area. Consequently,
291 for the KCMP tower site, we can consider the EC flux as representing the average flux from 600
292 km radius around the tall tower, a typical size of the footprint of tall tower concentration
293 measurement (10^6 km^2 ; Gloor et al., 2001).

294 The aggregated flux based on the STILT footprint was $-1.01 \mu\text{mol m}^{-2} \text{ s}^{-1}$ for the month of
295 September, 2009. In comparison, the EC flux during the same period was $-0.93 \mu\text{mol m}^{-2} \text{ s}^{-1}$, and
296 the FA flux with a 300 km and 600 km radius was $-1.04 \mu\text{mol m}^{-2} \text{ s}^{-1}$ and $-0.94 \mu\text{mol m}^{-2} \text{ s}^{-1}$,

297 respectively. The results again confirm that the land cover type around the tower was relatively
298 homogeneous at scales ranging from 200 to 600 km.

299 Consequently, we consider the tall-tower EC flux as a robust estimate of the regional flux and
300 used it to evaluate the performance of the EQ method. Two CO₂ fluxes were determined by EQ
301 method for each month, one using H₂O as a tracer (F_{EH} , calculated with ρW with Eq 3) and the
302 other using ρW in the NCEP reanalysis data (F_{EO} , calculated with ρW with Eq 4) (Figure 3).
303 Both F_{EH} and F_{EO} reproduced the seasonal pattern of the EC flux ($r > 0.85$, $p < 0.001$) but
304 significantly underestimated the magnitude of the flux in July. The F_{EH} and F_{EO} fluxes were only
305 31% and 48% of the EC flux in July.

306 **3.2 GHG concentration patterns**

307 During the intensive campaign, the CO₂ mixing ratio at the height of 200 m increased from 365.2
308 ppm during the first five days to 406.2 ppm during the last five days (Figure 4). The mixing ratio
309 changed from below that at the NWR (384.4 ppm) to above that at the NWR site, indicating a
310 transition of the landscape from a CO₂ sink to a source. This observation is consistent with the
311 seasonal pattern in the CO₂ flux shown in Figure 3.

312 The mean CH₄ mixing ratio during the observation period was 2.096 and 2.017 ppm at the
313 heights of 3 m and 200 m, respectively. The CH₄ mixing ratio at both heights was consistently
314 higher than the background mixing ratio at NWR (1.844 ppm), suggesting that the landscape
315 around the tall tower was a CH₄ source.

316 The N₂O mixing ratio during the observation period was also higher than that at NWR. The
317 average N₂O mixing ratio at the heights of 3 m and 200 m was 326.7 and 324.8 ppb, which were

318 4.0 ppb and 2.1 ppb higher than the value at NWR site (322.7 ppb), respectively, indicating that
319 the landscape was a N₂O source during the observation period.

320 **3.3 Regional CH₄ and N₂O fluxes**

321 We applied the EQ method in estimating the regional CH₄ and N₂O fluxes during the intensive
322 campaign. During this period ρW determined with three independent methods (using CO₂ and
323 H₂O tracers and the NCEP reanalysis data) was $-0.09 \pm 0.02 \text{ mol m}^{-2} \text{ s}^{-1}$ (mean \pm 1 standard
324 deviation of the three estimates) The CH₄ and N₂O fluxes were $16.0 \pm 3.1 \text{ nmol m}^{-2} \text{ s}^{-1}$ and 0.19
325 $\pm 0.04 \text{ nmol m}^{-2} \text{ s}^{-1}$, respectively.

326 In order to estimate an annual budget of CH₄ and N₂O with the EQ method, we need the CH₄ and
327 N₂O mixing ratio within and above the boundary layer for the whole year. Therefore, we
328 assumed that the seasonal pattern of the CH₄ and N₂O mixing ratios at the KCMP tower was
329 identical to the pattern at a nearby NOAA tall tower site (WBI in Iowa) (Andrews et al., 2013a;
330 Andrews et al., 2013b; Dlugokencky et al., 2013), and extrapolated the CH₄ and N₂O
331 concentration during the intensive campaign period to the whole year for 2009 according to the
332 seasonal pattern at the WBI site. The WBI site was chosen because it has similar land cover
333 types in its footprint as the KCMP tower site (Zhang et al., 2014). The CH₄ and N₂O mixing
334 ratios above the boundary layer were determined at the NWR site and ρW were determined by
335 the three methods (Equation 3-5) throughout the year 2009 (Figure S6). It follows that the annual
336 regional CH₄ and N₂O fluxes were $22.4 \pm 4.2 \text{ nmol m}^{-2} \text{ s}^{-1}$ and $0.49 \pm 0.09 \text{ nmol m}^{-2} \text{ s}^{-1}$,
337 respectively. The uncertainties of the annual fluxes were the result of the uncertainties in ρW . In
338 comparison, the annual CH₄ and N₂O fluxes at the WBI tower were $14.5 \text{ nmol m}^{-2} \text{ s}^{-1}$ and 0.32
339 $\text{nmol m}^{-2} \text{ s}^{-1}$ using the EQ method (Zhang, 2013). The impact of advection was considered as
340 negligible since there was no prevailing wind direction throughout the year of 2009.

341 4. Discussion

342 4.1 Annual carbon dioxide flux

343 Determining the annual CO₂ flux at the regional scale is challenging because the flux has both
344 diurnal and seasonal cycles and the magnitude of the annual average is substantially smaller than
345 the seasonal and diurnal variations. For example, in 2009, the tall tower's annual average EC
346 flux was $-0.35 \mu\text{mol m}^{-2} \text{s}^{-1}$ ($-131 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$), while the seasonal variation was about 6
347 $\mu\text{mol m}^{-2} \text{s}^{-1}$ and the diurnal variation during summer time was about $40 \mu\text{mol m}^{-2} \text{s}^{-1}$, about 16
348 and 113 times, respectively, higher than the annual average. So far, there is no single method that
349 can directly assess the regional CO₂ flux, because for all the available methods there are periods
350 or conditions where the underlying theory is not met or where the available data is limited to
351 truly capture the temporal and spatial variability. A small systematic bias in the daily and
352 monthly flux estimation, such as that caused by the data-screening and gap-filling approaches, is
353 significant for the annual average CO₂ flux, and it may result in opposite conclusions of whether
354 the landscape is a carbon source or a sink.

355 A number of studies have attempted to estimate the annual CO₂ flux in the vicinity of the Upper
356 Midwest USA (Table 2) (Davis et al., 2003; Bakwin et al., 2004; Helliker et al., 2004; Ricciuto
357 et al., 2008). Based on EC measurements on the LEF tall tower, which is about 260 km northeast
358 of the KCMP tower, Ricciuto et al. (2008) reported that the annual CO₂ flux was $120 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$
359 with a strong inter-annual variation from 1997 to 2004 ($140 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$). This result
360 was similar to Davis et al.'s (2003) result, but opposite to Helliker et al.'s (2004) EC flux for
361 2000, the latter of which is $-71 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$. The major difference is that Helliker et al.
362 (2004) did not gap-fill the data, and the reported CO₂ flux has excluded the periods when water
363 vapor flux was not available.

364 Our annual CO₂ flux was calculated as the average of monthly fluxes, and the monthly fluxes
365 were determined by the diurnal composite of available observations after u^* and storage term
366 screening. We performed a Monte Carlo simulation to assess the uncertainty associated with
367 missing data following Griffis et al. (2003). We randomly removed 30% of the data for each
368 month, and recorded the calculated monthly and annual fluxes following the same data
369 processing procedure. By repeating this simulation 5000 times, we determined the standard
370 deviation of the annual flux estimates. As a result, the uncertainty in the annual CO₂ flux due to
371 data gaps was $\pm 31 \text{ g C m}^{-2} \text{ yr}^{-1}$. In addition, the random errors in hourly averaged EC flux may
372 also significantly affect the annual budget. Assuming a 20% random error in hourly EC flux, the
373 resulting uncertainties in annual flux was $\pm 4 \text{ g C m}^{-2} \text{ yr}^{-1}$, about one magnitude lower than the
374 uncertainties from data gaps (Morgenstern et al., 2004). Considering the uncertainties from data
375 gaps and random errors, the 2009 CO₂ budget was $-131 \pm 35 \text{ g C m}^{-2} \text{ yr}^{-1}$, suggesting the region
376 around the tall tower was a carbon sink.

377 The uncertainty of the annual FA estimate was affected by the accuracy of the land cover
378 information, the carbon flux data for each land cover type, and anthropogenic emissions. We
379 assessed the uncertainties of the annual CO₂ flux from each land cover type with the method
380 used for tall tower EC flux (Table 2), and assume the uncertainty of anthropogenic emission was
381 10% (NRC, 2010). Without considering the uncertainties in land cover information, the annual
382 FA flux was $-130 \pm 13 \text{ g C m}^{-2} \text{ yr}^{-1}$. The accuracy of the Crop Data Layer was 85%-95% for
383 major crops (Boryan et al., 2011), therefore, we assume that the fraction of each land cover type
384 has an uncertainty up to 20%. By using a Monte Carlo simulation we estimated the uncertainty
385 of the annual FA estimate will increase to 34 g C m^{-2} . In other words, the FA annual flux ($-130 \pm$
386 $34 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$) showed very good agreement with the EC flux in 2009.

387 Although the CT monthly flux tracked the seasonal pattern of the EC and FA flux, the annual CT
388 flux was $-54 \text{ g C-CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$, considerably lower than EC and FA annual fluxes. This indicates
389 that the CT method performed reasonably well on reproducing the monthly fluxes, but that it
390 systematically underestimated the annual flux compared to the other methods.

391 Overall, the good agreement between EC (a top-down method) and FA (a bottom-up method)
392 provides strong evidence that the landscape around the tall tower was a carbon sink at the rate of
393 $-131 \pm 35 \text{ g C m}^{-2} \text{ yr}^{-1}$ in 2009.

394 **4.2 Uncertainties in CO₂ flux from the equilibrium method**

395 The EQ method provided good estimates of the CO₂ flux for each month except July, when the
396 regional CO₂ flux was the most negative (strong sink) during 2009. Excluding July, the
397 difference between the monthly EC flux and EQ flux in 2009 was $0.37 \pm 0.29 \mu\text{mol m}^{-2} \text{ s}^{-1}$, only
398 6% of the seasonal variation ($6 \mu\text{mol m}^{-2} \text{ s}^{-1}$).

399 The underestimation of the EQ flux might be attributed to uncertainties in ρW or concentration
400 difference at the top of boundary layer ($c_+ - c_m$), according to Equation 2. In July, the ρW was -
401 $0.17 \text{ mol m}^{-2} \text{ s}^{-1}$ (Eq 3) and $-0.26 \text{ mol m}^{-2} \text{ s}^{-1}$ (Eq 4) while the concentration difference was 9.03
402 ppm. To bring the equilibrium flux into agreement with the tall-tower EC flux ($-4.82 \mu\text{mol m}^{-2} \text{ s}^{-1}$
403 ¹ in July 2009), ρW would have to increase to $-0.53 \text{ mol m}^{-2} \text{ s}^{-1}$, which is much larger in
404 magnitude than $-0.26 \pm 0.09 \text{ mol m}^{-2} \text{ s}^{-1}$, the average July value for 2007 to 2011 obtained with
405 the NCEP reanalysis data. The $0.09 \text{ mol m}^{-2} \text{ s}^{-1}$ uncertainty in ρW (the standard deviation of July
406 ρW from 2007 to 2011) leads to $0.81 \mu\text{mol m}^{-2} \text{ s}^{-1}$ uncertainty in the monthly flux, about 17% of
407 the July flux. In addition, the monthly ρW values in 2007 to 2011 period were mostly within -
408 $0.18 \pm 0.08 \text{ mol m}^{-2} \text{ s}^{-1}$, and the maximum and minimum values were -0.05 and $-0.36 \text{ mol m}^{-2} \text{ s}^{-1}$.

409 Even the most negative value in the five-year period cannot fully explain the underestimation of
410 the EQ flux in July, indicating that the concentration difference at the top of boundary layer may
411 be underestimated.

412 Two sources of uncertainty exist in the concentration difference observed at the top of boundary
413 layer ($c_+ - c_m$). One is the accuracy of the CO₂ measurement at 200 m level of the KCMP tower
414 site and NWR background site, and the other is the assumption that those two concentrations are
415 the same as the concentration within and above the boundary layer. The CO₂ concentration at the
416 KCMP tower was calibrated with the NOAA-ESRL standard throughout the measurement, and
417 the measurement precision was 0.03 ppm. Meanwhile, the precision at NWR site was on the
418 order of 0.1 ppm (Helliker et al., 2004). As a result the first source of uncertainty led to about
419 0.04 $\mu\text{mol m}^{-2} \text{s}^{-1}$ uncertainties, which was not significant.

420 The second source of uncertainty can potentially lead to 1.55 $\mu\text{mol m}^{-2} \text{s}^{-1}$ uncertainties in the
421 July CO₂ flux. Even though the NWR site is only about 5° south of the KCMP tower, CO₂
422 concentration at the NWR site may be significantly different from the CO₂ concentration above
423 the boundary layer at the KCMP site, due to a large latitudinal CO₂ gradient (Denning et al.,
424 1995). To examine the uncertainties in using CO₂ concentration at NWR site as a proxy for c_+
425 we surveyed the following observation data that could be used as a proxy for c_+ in July 2009: 1)
426 the CO₂ concentration in the Marine Boundary Layer at the same latitude as the KCMP tower
427 was 382.47 ppm; 2) the average CO₂ concentration measured by aircraft (between 3000 m-4500
428 m) at three sites near the KCMP tower were 384.80 ± 4.55 ppm (Cooperative Global
429 Atmospheric Data Integration Project, 2014; Yi et al., 2004); 3) the average CO₂ concentrations
430 in the nearby background sites were 380.03 ppm (Cold Bay, Alaska, US) and 383.81 ppm
431 (Barrow, Alaska, US). The CO₂ concentration at the NWR site (386.01 ppm) is not significantly

432 different from the aircraft measurements and is lower than the rest of the concentrations with the
433 maximum difference of 5.98 ppm. Therefore, the CO₂ concentration at NWR site may
434 overestimate c_+ by up to 5.98 ppm and result in up to 1.55 $\mu\text{mol m}^{-2} \text{s}^{-1}$ uncertainty in the July
435 CO₂ flux. The uncertainties in other months in 2009 were examined and presented in the
436 supplementary materials (Section S4).

437 Bakwin et al. (2004) adjusted the CO₂ concentration at the 30 m height by increasing it by 2.5
438 ppm to estimate c_m in the summer. Since our CO₂ concentration was measured at 200 m, a much
439 higher level, the uncertainty in using 200 m concentration to estimate c_m should be much less
440 than 2.5 ppm. Direct measurement at the top of the boundary layer is needed to evaluate these
441 two important uncertainties.

442 Another more likely source for the significant underestimation in July is that horizontal
443 advection is not negligible when the prevailing winds align with a strong spatial CO₂ gradient.
444 With a tall tower observation network, Miles et al. (2012) reported that the CO₂ gradients
445 between the KCMP tower and other sites range from 0.3 to 2.1 ppm 100 km⁻¹ during the growing
446 season. The CarbonTracker 3-D CO₂ concentration product also shows large CO₂ depletion in
447 the upper Midwest Corn Belt during the growing season due to the strong CO₂ uptake by corn
448 plants. According to this product, the mean concentration of the 34 m – 1274 m air layer has an
449 averaged gradient of 0.8 ppm / (100 km) along the prevailing wind (from northwest) in July 2009
450 (Figure 5). Using a mean wind speed of 5.4 m s⁻¹ recorded on the tall tower and a boundary layer
451 depth of 1000 m (Yi et al., 2001), the resulting advection flux was -1.88 $\mu\text{mol m}^{-2} \text{s}^{-1}$, which is
452 comparable to the bias of the equilibrium method. In comparison, the EC flux was not as
453 sensitive to the advective influence. For instance, using the same spatial gradient data, the

454 advection flux at the 100 m level was only $-0.02 \mu\text{mol m}^{-2} \text{s}^{-1}$ according to the accumulated
455 concentration for the lowest two grid levels in the CarbonTracker product (34.5 m and 112 m).
456 Overall, the uncertainties in ρW , $(c_+ - c_m)$, and horizontal advection can potentially lead to
457 uncertainties in the July CO_2 flux estimate on the order of 0.81, 1.55, and $-1.88 \mu\text{mol m}^{-2} \text{s}^{-1}$
458 respectively, and they might be large enough to account for the discrepancy between EQ and EC
459 methods for July 2009 and the annual 2009 flux. Direct and accurate measurements of these
460 three terms using methods such as aircraft (Yi et al., 2004) or drones are needed to reduce these
461 uncertainties.

462 **4.3 Uncertainties in the CH_4 and N_2O fluxes**

463 Uncertainties in trace gas concentration measurements within and above the boundary layer can
464 lead to large uncertainties in the trace gas flux estimation. The averaged CH_4 concentrations
465 during the intensive campaign at the nearby background sites were 1.883 ppm (Cold Bay, Alaska,
466 US), 1.887 ppm (Barrow, Alaska, US), and 1.842 ppm (NWR, Colorado, US). The maximum
467 difference between NWR and the other background sites in North America was 0.045 ppm. If the
468 measurements at these other sites were used for the concentration above the boundary layer at
469 the tall tower site, the regional flux would decrease by up to $4.1 \text{ nmol m}^{-2} \text{s}^{-1}$, or about 30% of
470 the estimated EQ flux. In addition, the uncertainty in our CH_4 concentration measurement caused
471 by uncertainties in the calibration standard was 0.055 ppm, with a resulting uncertainty in the
472 flux of $\pm 5.0 \text{ nmol m}^{-2} \text{s}^{-1}$. The combined uncertainty range of the CH_4 flux is 6.9-21.0 nmol m^{-2}
473 s^{-1} .

474 The systematic bias in the trace gas measurement between the KCMP tower and NOAA
475 background sites was avoided in the other two independent boundary layer methods, which

476 depend on the relative concentration difference at 3 m and 200 m levels and the build-up (change
477 in concentration with time) concentration at night. Here, we used these two methods, a modified
478 Bowen ratio method (Werner et al., 2003) and a modified nocturnal boundary layer method
479 (Kelliher et al., 2002), to calculate the nighttime CH₄ flux for comparison with the equilibrium
480 estimate. The modified Bowen ratio method assumes that the vertical transport of a trace gas is
481 driven by eddy diffusion and that the diffusivity is the same for all scalar quantities. The
482 nocturnal boundary layer method uses CO₂ as a tracer and assumes that the build-up of CO₂ and
483 CH₄ near the land surface is caused by land surface emissions. The CH₄ fluxes from these two
484 methods were $14.8 \pm 10.3 \text{ nmol m}^{-2} \text{ s}^{-1}$ and $17.1 \pm 9.4 \text{ nmol m}^{-2} \text{ s}^{-1}$ (Zhang et al., 2013),
485 respectively. The results confirm that the CH₄ flux from the equilibrium method (16.0 nmol m^{-2}
486 s^{-1}) gave a reasonable estimation of the regional flux.

487 N₂O has a much more homogeneous background concentration than CH₄ and CO₂. The
488 differences between the background sites in the northern hemisphere were less than 0.5 ppb
489 during the intensive campaign. The N₂O measurements at the tall tower were calibrated against
490 NOAA-ESRL standards and, therefore, can be compared against the NOAA background sites.
491 The uncertainties in the background concentration will lead to a bias within $0.05 \text{ nmol m}^{-2} \text{ s}^{-1}$ for
492 the N₂O flux estimation, or 26% of the estimated N₂O flux ($0.19 \text{ nmol m}^{-2} \text{ s}^{-1}$). Applying the
493 modified Bowen ratio method and the modified nocturnal boundary layer method, we obtained a
494 regional nighttime N₂O flux of $1.09 \pm 0.56 \text{ nmol m}^{-2} \text{ s}^{-1}$ and $0.90 \pm 0.65 \text{ nmol m}^{-2} \text{ s}^{-1}$,
495 respectively, both of which were higher than the flux estimated from the equilibrium method. It
496 is possible that EQ method underestimated the regional N₂O flux because the advection was not
497 negligible during the intensive observation period, but it is not feasible to evaluate due to scarce
498 N₂O concentration measurements.

499 **4.4 Climate impact of the major GHG fluxes**

500 According to the KCMP tower measurement, the regional fluxes of three major greenhouse gases
501 in 2009 were -131 ± 35 g C-CO₂ m⁻² yr⁻¹, 8.50 ± 1.58 g C-CH₄ m⁻² yr⁻¹, and 0.43 ± 0.08 N-N₂O
502 m⁻² yr⁻¹. The global warming potential (GWP) over a 100-year time horizon for CO₂, CH₄, and
503 N₂O was -480 ± 128 , 283 ± 53 , and 205 ± 37 g CO₂ eq m⁻² yr⁻¹ respectively. The GWP for CO₂
504 was a result of anthropogenic emission (454 g CO₂ eq m⁻² yr⁻¹, according to the fossil fuel
505 emission prescribed in CarbonTracker product) and biological CO₂ uptake (934 g CO₂ eq m⁻² yr⁻¹)
506 for the region around the tall tower (Table 2). The total climate impact of the CH₄ and N₂O
507 emission offset about 30% and 22% of the biological CO₂ uptake and was comparable to the
508 anthropogenic CO₂ emission, indicating the important role of CH₄ and N₂O for the regional
509 GHG emission portfolio.

510 Considering all three major GHG fluxes, the landscape around the tall tower had a near neutral
511 impact on the climate in 2009 (7 ± 160 g CO₂ eq m⁻² yr⁻¹). This conclusion, however, did not
512 consider that the carbon fixed by crops will be harvested and some fraction will be transported
513 and emitted outside of the tall tower footprint. According to West et al. (2011), the harvested
514 biomass from the KCMP tower footprint is approximately 140 g C m⁻² yr⁻¹ (513 g CO₂ eq m⁻² yr⁻¹
515 ¹). In other words, the tall tower footprint likely has a warming impact on the climate when all
516 three major GHG fluxes and emission leakage (i.e. loss of carbon to the atmosphere from
517 harvested biomass) are considered.

518 **4.5 Comparison with bottom-up inventories**

519 EDGAR is a widely used anthropogenic GHG inventory for atmospheric research, having fine
520 spatial resolutions ($0.1^\circ \times 0.1^\circ$) (Jeong et al., 2012; Zhao et al., 2009). So far, only a few studies
521 have evaluated it with atmospheric observations. These studies indicate that EDGAR may have

522 significantly underestimate N₂O and CH₄ emission in North America by as much as three times
523 (Kort et al., 2008; Miller et al., 2012).

524 We first compared the CH₄ and N₂O fluxes at the KCMP tower during the intensive campaign
525 with EDGAR42 for the area within the 300 km radius around the tower, and found that the CH₄
526 flux was 5.8 times higher and the N₂O flux was 50% higher than the EDGAR42 values. In this
527 comparison, the EDGAR42 annual estimate was scaled to the emissions in September using its
528 seasonal factor (1.1 for September). Another comparison was carried out on the annual time
529 scale. The estimates of the annual CH₄ and N₂O fluxes based on the tall tower EQ measurement
530 were 6-9 times and 2-3 times higher than the EDGAR42 annual flux respectively.

531 The primary reason for the lower regional CH₄ flux from EDGAR42 is because it has excluded
532 natural sources of CH₄. Wetland is the major natural CH₄ source in this region. Although
533 wetlands account for less than 5% of the land around the tall tower, it is not negligible in the
534 regional CH₄ budgeting because CH₄ emissions from wetlands can be as high as 250 nmol m⁻² s⁻¹
535 in September (Bridgham et al., 2006). EDGAR42 may have also underestimated the CH₄
536 emission from anthropogenic sources, because it does not account for factors such as natural gas
537 leakage, and have low biases for the CH₄ emissions from agricultural activities (Mays et al.,
538 2009; Wunch et al., 2009; Ussiri et al., 2009).

539 We hypothesize that the lower N₂O flux in the EDGAR42 inventory is likely a result of the
540 underestimation of anthropogenic N₂O emission, since natural sources were not significant in the
541 region around the tall tower. A recent study on global N₂O emission from a natural ecosystems
542 suggests soil emissions in the upper-Midwest US is mostly around 0.10 kg N ha⁻¹ yr⁻¹ (0.01 nmol
543 m⁻² s⁻¹) (Zhuang et al., 2012), only 10% of the EDGAR42 anthropogenic emission.

544 In addition to EDGAR42, we compare the CH₄ and N₂O fluxes measured at the KCMP tower
545 with the GHG inventory developed by the EPA (EPA inventory), which was based on more
546 country-specific emission factors or models (e.g. N₂O from agriculture soil was simulated with a
547 biogeochemical model). The national CH₄ and N₂O emissions in EPA inventory were 12% and 35%
548 higher than the EDGAR42 inventory. However, we cannot directly compare the EPA inventory
549 with the top-down estimates for a region since the EPA inventory does not have spatial
550 distribution for all emission sectors. If we assume that the spatial distribution of the EPA
551 inventory is the same as the EDGAR42 inventory, the EPA inventory brings the bottom-up
552 estimates closer to the top-down EQ estimates. But the tall tower EQ estimates for regional CH₄
553 and N₂O emissions were still 5-8 times and 1-2 times higher than the EPA inventory.

554 **5. Conclusions**

555 The regional budget of CO₂, CH₄, and N₂O for the Upper Midwest, USA was quantified with
556 multiple top-down and bottom-up approaches. The four methods for the regional CO₂ flux (tall-
557 tower eddy covariance, CarbonTracker inverse modeling, flux aggregation, and the equilibrium
558 boundary layer method) produced similar seasonal patterns (linear correlation of the monthly
559 flux > 0.85, p < 0.001). However, discrepancies exist in the magnitude of the monthly and annual
560 fluxes. The CarbonTracker annual flux for 2009 (-54 g C-CO₂ m⁻² yr⁻¹) was much lower in
561 magnitude than the flux aggregation estimate (- 130 ± 37 g C-CO₂ m⁻² yr⁻¹) and that measured
562 with eddy covariance (-131 ± 40 g C-CO₂ m⁻² yr⁻¹). The equilibrium method significantly
563 underestimated the July uptake by 52-69% in comparison to eddy covariance. The
564 underestimation cannot be fully explained by the bias in ρW or concentration difference at the
565 top of the boundary layer, and we suggest that the large spatial gradient along the prevailing
566 wind in July 2009 was a main contributor to the underestimation.

567 The CH₄ and N₂O regional fluxes estimated from equilibrium method during the intensive
568 campaign (DOY 243 – 269, 2009) were 16.0 ± 3.1 and 0.19 ± 0.04 nmol m⁻² s⁻¹, respectively,
569 and were 5.8 times and 50% higher than in the EDGAR42 inventory. The annual CH₄ and N₂O
570 fluxes also suggest significant underestimation by the EDGAR42 inventory and the EPA
571 inventory.

572 Considering the global warming potential on a 100-year time scale, the CH₄ and N₂O emissions
573 from the landscape were comparable to the anthropogenic CO₂. The landscape appeared to have
574 a near neutral impact on climate when all three major GHGs were considered. Our results
575 confirm that for this agriculture dominated landscape, climate change mitigation should include
576 CH₄ and N₂O emissions.

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870 **Tables**

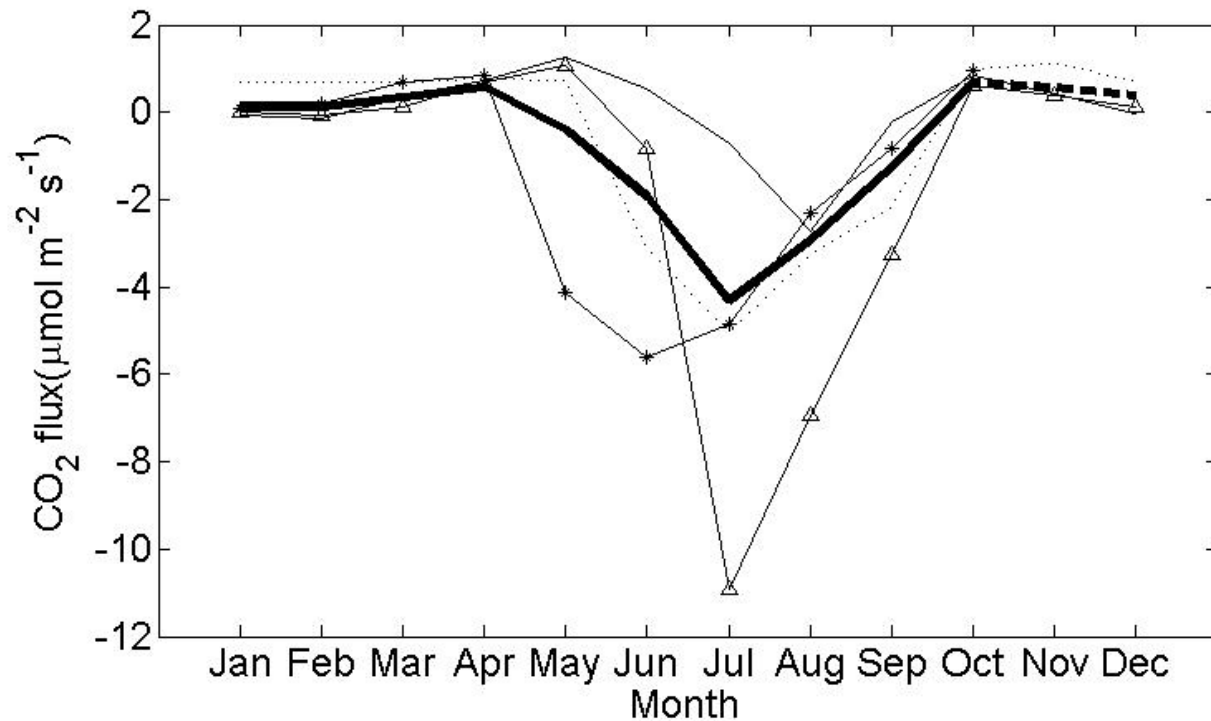
871 Table 1. Correlation coefficient and NSE (Nash-Sutcliffe efficiency) between EC flux and the other two methods. CT denotes
 872 CarbonTracker, and FA denotes the flux aggregation method.

Distance		5 km	10 km	20 km	50 km	100 km	200 km	300 km	600 km
CT	NSE	NA	NA	NA	NA	0.41	0.82	0.92	0.97
	r	NA	NA	NA	NA	0.96	0.97	0.98	0.99
FA	NSE	0.48	0.49	0.37	0.23	0.58	0.90	0.95	0.96
	r	0.98	0.98	0.98	0.98	0.98	0.98	0.98	0.98

873

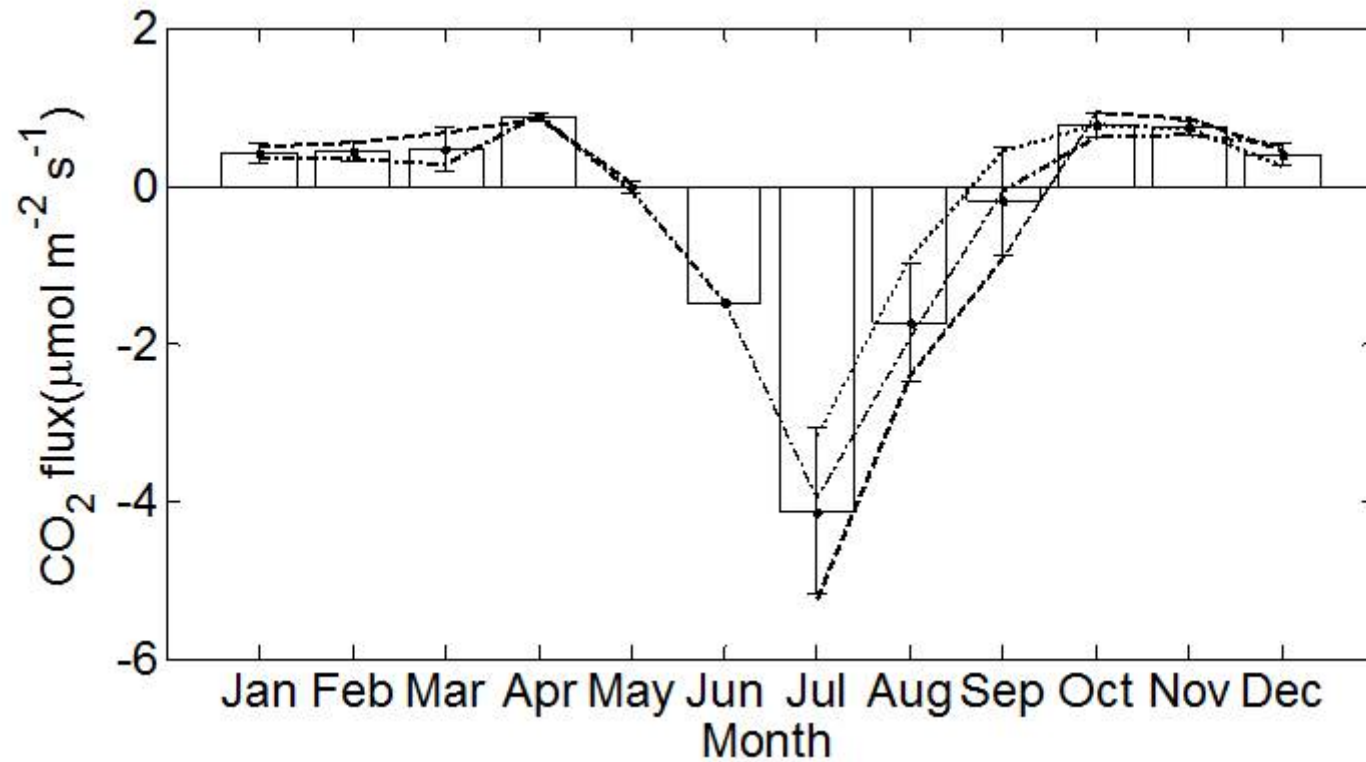
874 Table 2. A summary of annual NEE estimated from different methods. NEE in the “Reference” column is from the study in Midwest
 875 US in recent years. Negative fluxes indicate carbon sink from the atmosphere and positive fluxes indicate carbon release to the
 876 atmosphere.

	This study (g C-CO ₂ m ⁻² yr ⁻¹)	Reference (g C-CO ₂ m ⁻² yr ⁻¹)
Tall tower Eddy Covariance	-131 ± 35	120 ± 140 (1997-2004) (Ricciuto et al., 2010) 16 ± 19 (1997) (Davis et al., 2003) -71 (2000) (Helliker et al., 2004) 16 (1997) (Bakwin et al., 2004)
CarbonTracker	-54 ± 12	-58 (2000-2006) (Desai et al., 2010)
Equilibrium	46-74	-110 ± 14 (1997-2006) (Desai et al., 2010) -38 (2000) (Helliker et al., 2004) 79 (1997) (Bakwin et al., 2004)
Flux Aggregation	-130 ± 34	
Corn	-599 ± 26	-466 ± 38 (2004-2007) (Hernandez-Ramirez et al., 2011) -576 ± 101 (1997-2002) (Hollinger et al., 2005)
Soybean	10 ± 18	-13 ± 39 (2004-2007) (Hernandez-Ramirez et al., 2011) -32 ± 161 (1997-2002) (Hollinger et al., 2005)
Grassland	-411 ± 10	-148 ± 116 (1997-1999) (Suyker et al., 2003)
Forest	-227 ± 14	-137 ± 49 (1999-2001) (Schmid et al., 2003)
Fossil fuel	124 ± 12	
Other methods		
Interannual Flux Tower Upscaling Experiment		-321 ± 13 (1997-2006) (Desai et al., 2010)
Mesoscale inverse modeling		-183 ± 35 (1997-2006) (Lauvaux et al., 2012)



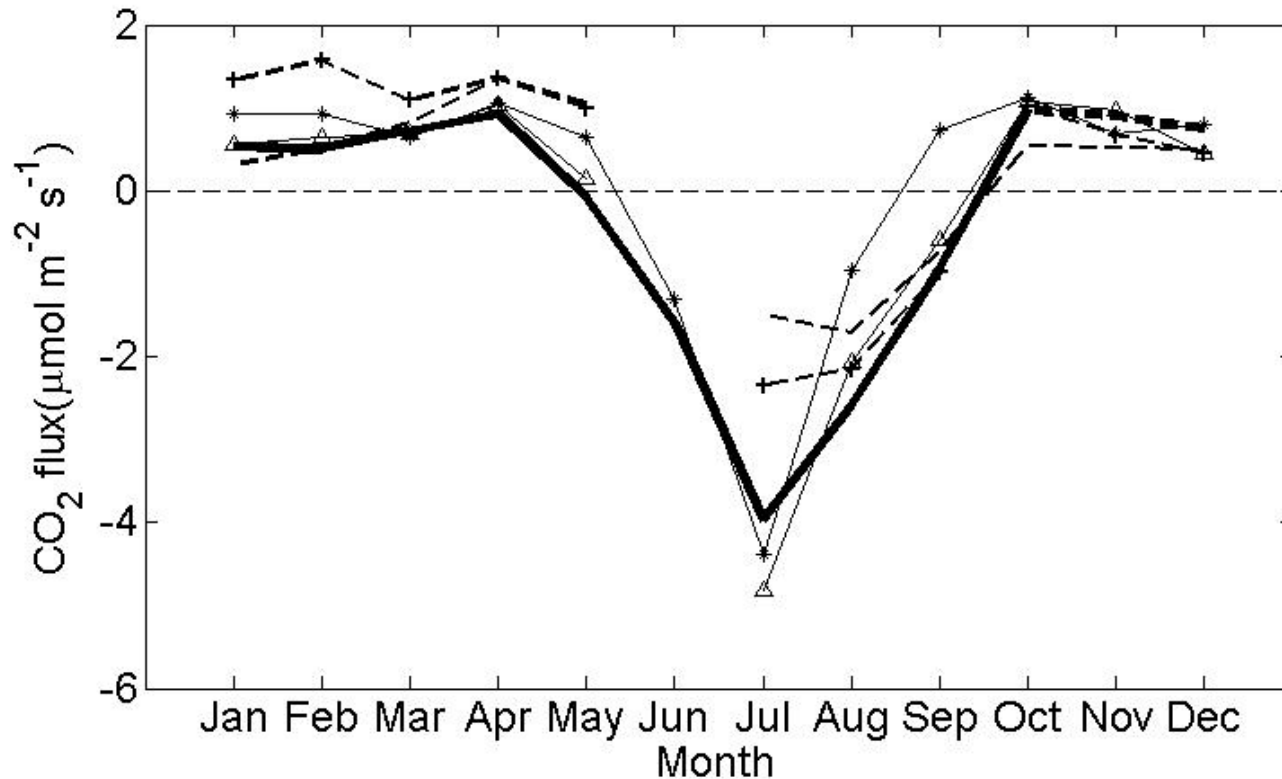
879

880 Figure 1. The biogenic CO₂ flux (thick solid line) from the landscape around the tall tower, calculated from monthly averages of CO₂
881 flux from major land cover types. The fluxes for corn (solid line with triangles) and soybean (thin solid line) were measured with eddy
882 covariance tower at the G21 and G19 site near the tall tower in Minnesota. The fluxes for forest (dotted line) and grassland (solid line
883 with stars) were from UMBS and USIB2 AmeriFlux sites in North America.



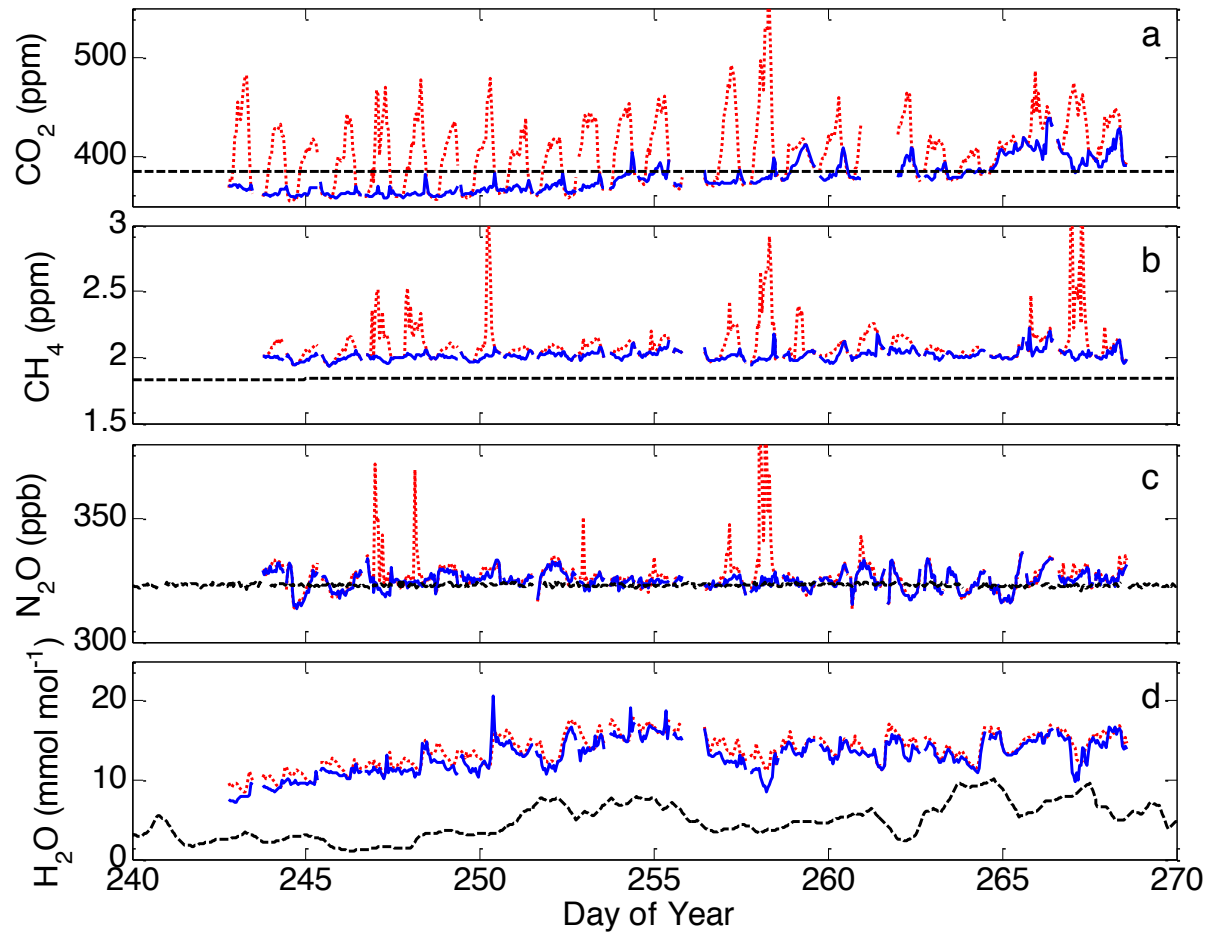
884

885 Figure 2. Monthly averages of CO₂ flux in 2007 (dotted line), 2008 (dot-dashed line), and 2009 (dashed line) measured with EC on the
 886 tall tower. White bars are the mean monthly value from the available data during the three-year observation period. Error bars on the
 887 top of white bars show the lower and upper boundary of the three-year measurements for each month.



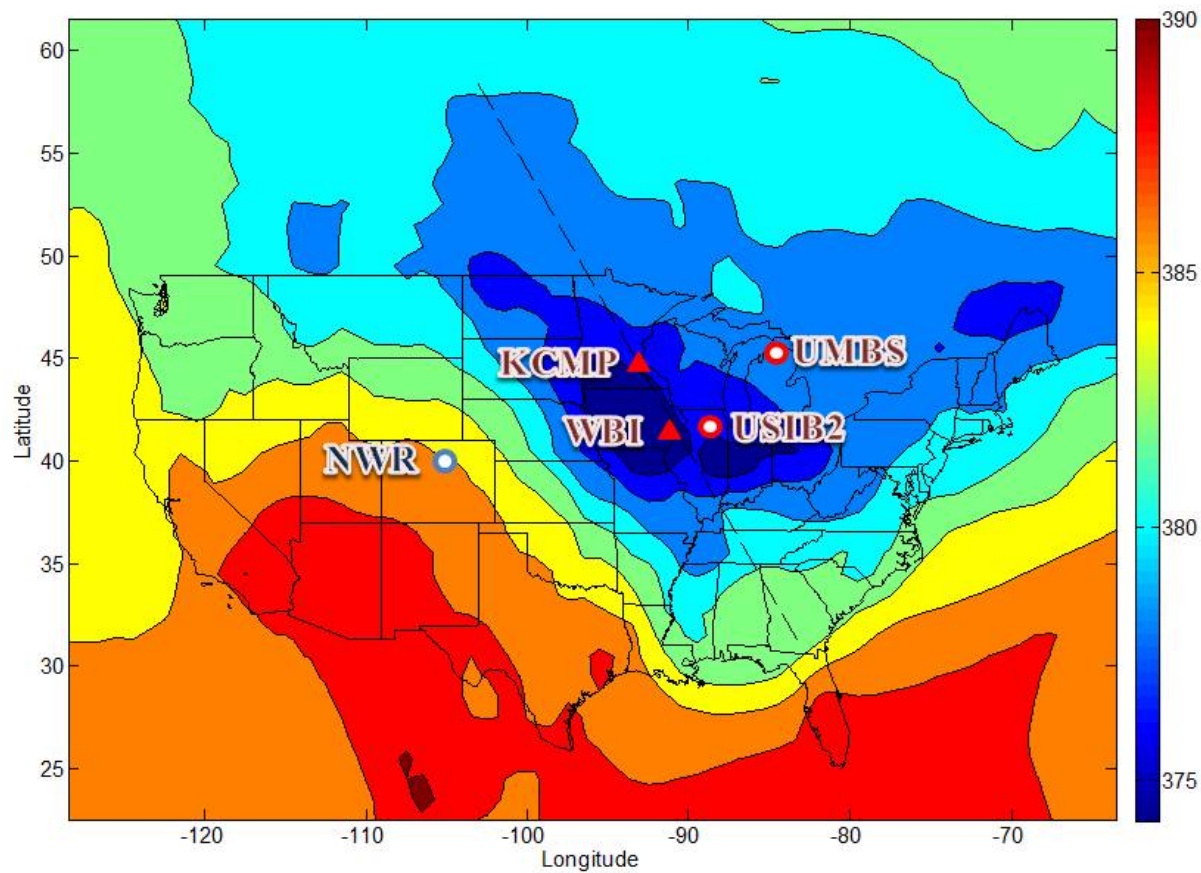
888

889 Figure 3. Monthly CO₂ flux in 2009 estimated with flux aggregation (FA, thick solid line), eddy covariance method (F_{EC}, line with
 890 triangles), CarbonTracker (F_{CT}, line with stars), equilibrium method with H₂O as a tracer (F_{EH}, thin dashed line), and equilibrium
 891 method using NCEP reanalysis data (F_{EO}, thin dashed line with cross). The FA flux ended in October because the grassland data was
 892 missing in November and December. The missing FA flux was determined by assuming the missing grassland flux in November and
 893 December was the same as October flux (thick dashed line).



894

895 Figure 4. Hourly averages of CO₂ (a), CH₄ (b), N₂O (c), and H₂O (d) mixing ratio during the observation period from DOY 243 to
 896 DOY 269, 2009. Blue solid line— mixing ratio on 200 m. Red dotted line— mixing ratio on 3 m. Black dashed line— mixing ratio at
 897 Niwot Ridge site.



898

899 Figure 5. CO₂ concentration averaged from land surface to 1274.1 m according to the CarbonTracker 3D CO₂ concentration product in
 900 July, 2009. Dashed line — prevailing wind direction in July from northwest to southeast. Red triangle — the KCMP tower site and
 901 WBI tower observatory operated by NOAA. Red circle— Ameriflux sites. Blue circle — background observation site. The color
 902 scale is the CO₂ concentration in ppm. The resolution of the concentration data is 1° by 1°.