

# 1 **Background Heterogeneity and Other Uncertainties in** 2 **Estimating Urban Methane Flux: Results from the** 3 **Indianapolis Flux (INFLUX) Experiment**

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## 13 14 **Abstract**

15 As natural gas extraction and use continues to increase, the need to quantify emissions of  
16 methane (CH<sub>4</sub>), a powerful greenhouse gas, has grown. Large discrepancies in Indianapolis CH<sub>4</sub>  
17 emissions have been observed when comparing inventory, aircraft mass-balance, and tower  
18 inverse modeling estimates. Four years of continuous CH<sub>4</sub> mole fraction observations from a  
19 network of nine towers as a part of the Indianapolis Flux Experiment (INFLUX) are utilized to  
20 investigate four possible reasons for the abovementioned inconsistencies: (1) differences in  
21 definition of the city domain, (2) a highly temporally variable and spatially non-uniform CH<sub>4</sub>  
22 background, (3) temporal variability in CH<sub>4</sub> emissions, and (4) the presence of unknown CH<sub>4</sub>  
23 sources. Reducing the Indianapolis urban domain size to be consistent with the inventory  
24 domain size decreases the CH<sub>4</sub> emission estimation of the inverse modeling methodology by  
25 about 35% and thereby lessens the discrepancy by bringing total city flux within an error range  
26 of one of the inventories. Nevertheless, the inverse modeling estimate still remains about 40%  
27 higher than the inventory value. Hourly urban background CH<sub>4</sub> mole fractions are shown to be  
28 heterogeneous and temporally variable. Variability in a single point background mole fractions  
29 observed at any given moment could be up to about 50 ppb depending on a wind direction, but

30 decreases substantially when averaged over multiple days. Statistically significant, long-term  
31 biases in background mole fractions of 2-5 ppb are found from single point observations from  
32 most wind directions. Boundary layer budget estimates suggest that Indianapolis CH<sub>4</sub> emissions  
33 did not change significantly when comparing 2014 to 2016. However, it appears that CH<sub>4</sub>  
34 emissions may follow a diurnal cycle with daytime emissions (12-16 LST) approximately twice  
35 as large as nighttime emissions (20-5 LST). No significant unknown CH<sub>4</sub> sources are found.  
36 The data from the towers suggest that the strongest CH<sub>4</sub> source in Indianapolis is South Side  
37 Landfill. Other sources, such as leaks from the natural gas (NG) distribution system, are  
38 localized and transient, and do not appear to be a consistently large source of CH<sub>4</sub> emissions in  
39 Indianapolis. However, some uncertainty regarding occasional significant CH<sub>4</sub> leaks from NG  
40 distribution exists. Long-term averaging, spatially-extensive upwind mole fraction observations,  
41 mesoscale atmospheric modeling of the regional emissions environment, and careful treatment of  
42 the times of day and areal representation of emission estimates are recommended for precise and  
43 accurate quantification of urban CH<sub>4</sub> emissions.

44

## 45 **1 Introduction**

46 From the beginning of the Industrial Revolution to 2011, atmospheric methane (CH<sub>4</sub>) mole  
47 fractions increased by a factor of 2.5 due to anthropogenic processes such as fossil fuel  
48 production, waste management, and agricultural activities (Ciais et al., 2013). The increase in  
49 CH<sub>4</sub> is a concern as it is a potent greenhouse gas (GHG) with a global warming potential 28-34  
50 times greater than that of CO<sub>2</sub> over a period of 100 years (Myhre et al., 2013). The magnitudes  
51 of component CH<sub>4</sub> sources, and the causes of variability in the global CH<sub>4</sub> budget, however, are  
52 not well understood although there is some evidence that biogenic emissions may play an

53 important role in the recent CH<sub>4</sub> increases (Nisbet et al., 2016; Saunio et al., 2016). Improved  
54 understanding of CH<sub>4</sub> emissions is needed (National Academies of Sciences and Medicine,  
55 2018).

56 In particular, the estimates of continental U.S. anthropogenic CH<sub>4</sub> emissions disagree.  
57 Inventories from Environment Protection Agency (EPA) and Emissions Database for Global  
58 Atmospheric Research (EDGAR) in 2008 reported emission values of 19.6 and 22.1 TgC y<sup>-1</sup>  
59 (Miller et al., 2013). However, top-down methodologies using aircraft and inverse modeling  
60 framework found emission values of 32.4 ± 4.5 TgC y<sup>-1</sup> for 2004 and 33.4 ± 1.4 TgC y<sup>-1</sup> for  
61 2007-2008 respectively (Kort et al., 2008; Miller et al., 2013). Underestimation of natural gas  
62 (NG) production and agricultural sources are possible reasons for this disagreement (Miller et al.,  
63 2013; Brandt et al., 2014; Jeong et al., 2014). Efforts to reconcile GHGs emissions estimates  
64 using atmospheric methods and inventory assessment have sometimes succeeded (Schuh et al.,  
65 2013; Zavala-Araiza et al., 2015; Turnbull et al., 2019) when careful attention is given to the  
66 details of each method, and targeted atmospheric data are available. A recent synthesis of  
67 emissions from the U.S. NG supply chain demonstrated similar success and concluded that  
68 current inventory estimates of emissions from U.S. NG production are too low and that emission  
69 from NG distribution is one of the greatest remaining sources of uncertainty in the NG supply  
70 chain (Alvarez et al., 2018).

71 Due to the uncertainties in CH<sub>4</sub> emissions from NG distribution it is natural that urban  
72 emissions are of interest as well. For example, studies indicate that ~60-100% of Boston CH<sub>4</sub>  
73 emissions are attributable to the NG distribution system (McKain et al., 2015; Hendrick et al.,  
74 2016). Recent studies of urban CH<sub>4</sub> emissions in California indicate that the California Air  
75 Resources Board (CARB) inventory tends to underestimate the actual CH<sub>4</sub> urban fluxes possibly

76 due to fugitive emissions that result from the NG infrastructures common to the urban  
77 environments (Wunch et al., 2009; Jeong et al., 2016; Jeong et al., 2017). The accuracy and  
78 precision of atmospheric estimates of urban CH<sub>4</sub> emissions are limited by available atmospheric  
79 observations (Townsend-Small et al., 2012), potential source magnitude variability with time  
80 (Jackson et al., 2014; Lamb et al., 2016), errors in atmospheric transport modeling (Hendrick et  
81 al., 2016; Deng et al., 2017; Sarmiento et al., 2017), and complexity in atmospheric background  
82 conditions (Cambaliza et al., 2014; Karion et al., 2015; Heimburger et al., 2017). In this work,  
83 detailed analysis of urban CH<sub>4</sub> mole fractions is performed for the city of Indianapolis to better  
84 understand the aforementioned uncertainties of urban CH<sub>4</sub> emissions.

85         The Indianapolis Flux Experiment (INFLUX; Davis et al., 2017) is a testbed for  
86 improving quantification of urban GHGs emissions and their variability in space and time.  
87 INFLUX (<http://influx.psu.edu>) is located in Indianapolis partly because of its isolation from  
88 other urban centers and the flat Midwestern terrain. It includes a very dense GHGs monitoring  
89 network, comprised of irregular insitu aircraft measurements (Heimburger et al., 2017;  
90 Cambaliza et al., 2014), continuous in situ observations from communications towers using  
91 cavity ring-down spectroscopy (Richardson et al., 2017; Miles et al., 2017), and automated flask  
92 sampling systems for quantification of a wide variety of trace gases (Turnbull et al., 2015).  
93 Meteorological sensors include a Doppler lidar providing continuous boundary layer depth and  
94 wind profiles, and tower-based eddy covariance measurements of the fluxes of momentum,  
95 sensible and latent heat (Sarmiento et al., 2017). The network is designed for emissions  
96 estimates using top-down methods such as tower-based inverse modeling (Lauvaux et al., 2016)  
97 and aircraft mass balance estimates (Cambaliza et al., 2015).



98 Lamb et al. (2016) compared Indianapolis CH<sub>4</sub> emissions estimates from a variety of  
99 approaches, which are inventory, aircraft mass balances, and inverse modeling. The results  
100 revealed large mean differences among the city fluxes estimated from these methods (Fig. 1). In  
101 general, the inventory methods arrived at lower estimates of emissions compared to the  
102 atmospheric, or top-down approaches. CH<sub>4</sub> fluxes calculated using the aircraft mass balance  
103 technique varied considerably between flights, more than would be expected from propagation of  
104 errors of the component measurements (Cambaliza et al., 2014; Lamb et al., 2016). The  
105 atmospheric inverse estimate was significantly higher than the inventory and some of the  
106 aircraft-derived values.

107 Biogenic emissions from the city are dominated by a landfill close to downtown, and  
108 these emissions are thought to be fairly well known (GHG reporting program). Although  
109 evidence of possible variability in landfill emissions exists from Cambaliza et al. (2015) study  
110 that used aircraft mass balance on five different occasions to calculate CH<sub>4</sub> flux from this  
111 landfill. Uncertainty in total city emissions is mainly driven by the uncertainty in thermogenic  
112 emissions, which are hypothesized to emerge largely from the NG distribution system (Mays et  
113 al., 2009; Cambaliza et al., 2015; Lamb et al., 2016). This uncertainty has not yet been resolved.  
114 In this study, we explore potential explanations for the discrepancies in CH<sub>4</sub> emissions estimates  
115 from Indianapolis and posit methods and recommendations for the study of CH<sub>4</sub> emissions from  
116 other urban centers.

117 We examine four different potential explanations for the CH<sub>4</sub> flux discrepancies reported  
118 in Lamb et al. (2016): (1) inconsistent geographic boundaries, (2) heterogeneity in the urban-  
119 scale CH<sub>4</sub> background, (3) temporal variability in urban emissions, and (4) CH<sub>4</sub> sources that are  
120 not accounted for in the inventories. Well-calibrated CH<sub>4</sub> sensors on the INFLUX tower network

121 (Miles et al., 2017) collected continuous CH<sub>4</sub> observations from 2013 to 2016 and provide a  
122 unique opportunity to explore these issues.

123

## 124 **2 Methods**

125

### 126 **2.1 Experimental site**

127 This study uses data from a tower-based GHG observational network located in the city and  
128 surrounding suburbs of Indianapolis, Indiana in the Midwestern U.S. Prior studies have used  
129 varying definitions for the region of Indianapolis (Cambaliza et al., 2015, Lamb et al., 2016). In  
130 this work, we follow Gurney et al. (2012) and define Indianapolis as the area of Marion County.  
131 The flat terrain of the region simplifies interpretation of the atmospheric transport. The land-  
132 surface heterogeneity inherent in the urban environment (building roughness, spatial variations in  
133 the surface energy balance) does have a modest influence on the flow within the city and the  
134 boundary layer depth difference between the urban and rural areas (Sarmiento et al., 2017).

135 Figure 2 shows two domains that have been used for the evaluation of Indianapolis CH<sub>4</sub>  
136 emissions (Lamb et al., 2016; Lauvaux et al., 2016). The first domain is the whole area shown in  
137 the figure enclosing both Indianapolis and places that lie outside of its boundaries. This domain  
138 has been used for the inversion performed in Lamb et al. (2016). The second domain is Marion  
139 County outlined with a green dashed line. It is assumed here that this domain is much more  
140 representative of the actual Indianapolis municipal boundaries as this area encompasses the  
141 majority of the urban development associated with the city of Indianapolis (Gurney et al., 2012).  
142 The larger domain has three additional landfills that based on the EPA gridded inventory  
143 (Maasackers et al., 2016) increase Indianapolis CH<sub>4</sub> emissions by about 50% when compared to

144 the smaller domain. The inversion explained in Lamb et al. (2016) has been rerun for two of the  
145 domains mentioned above and the results (Fig. 1) have been reexamined.

146

## 147 **2.2 INFLUX tower network**

148 The continuous GHG measurements from INFLUX are described in detail in Richardson et al.  
149 (2017). The measurements were made using wavelength-scanned cavity ring down  
150 spectrometers (CRDS, Picarro, Inc., models G2301, G2302, G2401, and G1301), installed at the  
151 base of existing communications towers, with sampling tubes secured as high as possible on each  
152 tower (39 – 136 m above ground level (AGL); Miles et al., 2017). A few towers also included  
153 measurements at 10 m AGL and one or two intermediate levels. While INFLUX tower in-situ  
154 measurements began in September 2010, here we focus on the CH<sub>4</sub> measurements from 2013 –  
155 2016. From June through December 2012, there were two or three towers with operational CH<sub>4</sub>  
156 measurements. By July 2013, five towers included measurements of CH<sub>4</sub>, and throughout the  
157 majority of the years 2015 – 2016 there were eight INFLUX towers with CH<sub>4</sub> measurements  
158 (Fig. 3). Flask to in-situ comparisons and round-robin style testing indicated compatibility  
159 across the tower network of 0.6 ppb CH<sub>4</sub> (Richardson et al., 2017). In this study we use hourly  
160 means of CH<sub>4</sub>, which were reported on the WMO X2004A scale.

161

## 162 **2.3 Meteorological data**

163 Wind data was measured at the Indianapolis International Airport (KIND), Eagle Creek Airpark  
164 (KEYE), and Shelbyville Municipal Airport (KGEZ). The data used are hourly values from the  
165 Integrated Surface Dataset (ISD) (<https://www.ncdc.noaa.gov/isd>) and 5-minute values directly  
166 from the Automated Surface Observing System (ASOS). A complete description of ASOS

167 stations is available at <https://www.weather.gov/media/asos/aum-toc.pdf>. The accuracy of the  
168 wind speed is  $\pm 1$  m/s or 5% (whichever is greater) and the accuracy of the wind direction is 5  
169 degrees when the wind speed is  $\geq 2.6$  m/s. The anemometers are located at about 10 meters  
170 AGL. The wind data reported in ISD are given for a single point in time recorded within the last  
171 10 minutes of an hour and are closest to the value at the top of the hour.

172 The planetary boundary layer height (BLH) was determined from a Doppler lidar  
173 deployed in Lawrence, Indiana about 15 km to the northeast of downtown. The lidar is a Halo  
174 Streamline unit, which was upgraded to have extended range capabilities in January 2016. The  
175 lidar continuously performs a sequence of conical, vertical-slice, and staring scans to measure  
176 profiles of the mean wind, turbulence, and relative aerosol backscatter. All of these  
177 measurements are combined using a fuzzy-logic technique to automatically determine the BLH  
178 continuously every 20-min (Bonin et al., 2018). The BLH is primarily determined from the  
179 turbulence measurements, but the wind and aerosol profiles are also used to refine the BLH  
180 estimate. The BLHs are assigned quality-control flags that can be used to identify times when  
181 the BLH is unreliable, such as when the air is exceptionally clean, the BLH is below a minimum  
182 detectable height, or clouds and fog that attenuate the lidar signal exist. Additional details about  
183 the algorithm and the lidar operation for the INFLUX project are provided in Bonin et al. (2018).  
184 Doppler lidar measurements are available at <https://www.esrl.noaa.gov/csd/projects/influx/>.

185

## 186 **2.4 Urban methane background**

187 Both aircraft mass balance and inverse modeling methodologies rely on an accurate estimation of  
188 the urban CH<sub>4</sub> enhancement relative to the urban CH<sub>4</sub> background in order to produce a reliable

189 flux estimate (Cambaliza et al., 2014; Lamb et al., 2016). The CH<sub>4</sub> mole fraction enhancement is  
190 defined as,

$$C_{enhancement} = C_{downwind} - C_{bg} \quad (1)$$

191 where  $C_{downwind}$  is the CH<sub>4</sub> mole fraction measured downwind of a source and  $C_{bg}$  is the CH<sub>4</sub>  
192 background mole fraction, which can be measured upwind of the source, but this is not  
193 necessary. Background, as defined in this body of literature, is a mole fraction measurement that  
194 does not contain the influence of the source of interest, but which is assumed to accurately  
195 represent mole fractions that are upwind of the source of interest and measured simultaneously  
196 with the downwind mole fractions.

197 Aircraft mass balance studies at Indianapolis mentioned in this article used two main  
198 methods to determine a background value. The first method calculates an average of the aircraft  
199 transect edges that lie outside of the city domain (Cambaliza et al., 2014). In the second  
200 approach, a horizontally varying background is introduced by linearly interpolating median  
201 background values of each of the transect edges (Heimbürger et al., 2017). In theory there is also  
202 a third method that uses an upwind transect as a background field, but in the studies above it was  
203 assumed that the edges are representative of an upwind flow. In the case of an inversion, it is  
204 common to pick a tower that is located generally away from urban sources and has on average  
205 the smallest overall enhancement (Lavaux et al., 2016). Because choosing the background  
206 involves a degree of subjectivity (Heimbürger et al., 2017) we consider how these choices may  
207 influence emission estimates and introduce error, both random and systematic, using data from  
208 the INFLUX tower network.

209 Using tower network data from November 2014 through the end of 2016, two CH<sub>4</sub>  
210 background fields are generated for the city of Indianapolis based on two different sets of

211 criteria. The notion is based on the fact that a choice of background is currently rather arbitrary  
212 in the literature (Heimburger et al., 2017) and at every point in time it is possible to choose  
213 multiple background values that are equally acceptable for the flux estimation. In our case both  
214 approaches identify a tower suitable to serve as a background for each of the eight wind  
215 directions (N, NE, E, SE, S, SW, W, NW), where an arc of  $45^\circ$  represents a direction (e.g. winds  
216 from N are between  $337.5^\circ$  and  $22.5^\circ$ ). Estimating background for different wind directions is  
217 implemented to more accurately represent upwind flow that is hopefully not contaminated by  
218 local sources.

219 Criterion 1 corresponds to a typical choice of a background in a case of tower inversion  
220 and is based on the concept that the lowest  $\text{CH}_4$  mole fraction measured at any given time is not  
221 affected by the city sources and therefore is a viable approximation of the background  $\text{CH}_4$  mole  
222 fractions outside of the city (Miles et al., 2017; Lauvaux et al., 2016). Given this assumption, the  
223 tower with the lowest median of the  $\text{CH}_4$  enhancement distribution (calculated by assuming the  
224 lowest measurement among all towers at a given hour as a background) for each of the wind  
225 directions over the November 2014 through December 2016 time period is chosen as a  
226 background site (Miles et al., 2017). Criterion 2 requires that the tower is outside of Marion  
227 County (outside of the city boundaries) and is not downwind of any known regional  $\text{CH}_4$  source  
228 (Fig. 2). For some wind directions, there are multiple towers that could qualify as a background;  
229 we pick towers in such a manner that they are different for each criterion given a wind direction  
230 in order to calculate the error associated with the use of different but acceptable backgrounds.  
231 The towers used for both criteria and for each of the eight wind directions are displayed in Table  
232 1. Quantifying differences between these two backgrounds allows for an opportunity to better

233 understand the degree of uncertainty that exists in the atmospheric CH<sub>4</sub> background at  
234 Indianapolis.

235 To make the comparison as uniform as possible only data from 12-16 LST are utilized  
236 (all hours are inclusive) when the boundary layer is typically well-mixed (Bakwin et al., 1998).  
237 A lag 1 autocorrelation is found between 12-16 LST hours, i.e. the hourly afternoon data are  
238 correlated to the next hour, but the correlation is not significant for samples separated by two  
239 hours or more. Therefore, hours 13 and 15 LST are eliminated to satisfy the independence  
240 assumption for hourly samples. Furthermore, we make an assumption that the data satisfy steady  
241 state conditions. If the difference between consecutive hourly wind directions exceeds 30  
242 degrees or the difference between hours 16 and 12 LST exceeds 40 degrees, the day is  
243 eliminated. Days with average wind speeds below 2 m/s are also eliminated due to slow  
244 transport (the transit time from tower 1 to tower 8 is about 7 hours at a wind speed of 2 m/s).

245

## 246 **2.5 Frequency and bivariate polar plots**

247 Frequency and bivariate polar plots are used in this work to gain more knowledge regarding CH<sub>4</sub>  
248 background variability based on criteria 1 and 2, and to identify sources located within the city.  
249 To generate these polar plots, we use the *openair* package (from R programming language)  
250 created specifically for air quality data analysis (Carslaw and Ropkins, 2012). Bivariate and  
251 frequency polar plots indicate the variability of a pollutant concentration at a receptor (such as an  
252 observational tower) as a function of wind speed and wind direction, preferably measured at the  
253 location of the receptor or within several kilometers of the receptor. The frequency polar plot is  
254 generated by partitioning the CH<sub>4</sub> hourly data into the wind speed and direction bins of 1 m s<sup>-1</sup>  
255 and 10° respectively. To generate bivariate polar plots, wind components *u* and *v* are calculated

256 for hourly CH<sub>4</sub> mole fraction values, which are fitted to a surface using a Generalized Additive  
257 Model (GAM) framework in the following way,

$$\sqrt{C} = \beta + s(u, v) + \epsilon \quad (2)$$

258 where  $C$  is the CH<sub>4</sub> mole fraction transformed by a square root to improve model diagnostics  
259 such as a distribution of residuals,  $\beta$  is mean of the response,  $s$  is the isotropic smoothing  
260 function of the wind components  $u$  and  $v$ , and  $\epsilon$  is the residual. For more details on the model  
261 see Carslaw and Beevers (2013).

262

## 263 **2.6 Temporal variability and approximate flux estimation**

264 Temporal variability may play an important role in the quantification of urban CH<sub>4</sub> emissions.  
265 Lamb et al., (2016) suggested that temporal variability might partially explain the differences  
266 among CH<sub>4</sub> flux estimates shown in Figure 1. If temporal variability of CH<sub>4</sub> emissions exists  
267 within the city, disagreements in the CH<sub>4</sub> flux between studies could be attributed to differences  
268 in their sampling period. Because the INFLUX tower data at Indianapolis contain measurements  
269 at all hours of the day over multiple years, we can utilize this dataset to better understand the  
270 temporal variability in methane emissions in the city.

271 We apply a simplified atmospheric boundary layer budget, not to estimate precisely the  
272 actual city emissions, but rather to evaluate temporal variability of the emissions. We begin by  
273 assuming CH<sub>4</sub> emissions  $Q_a$  (mass per unit time per unit area) are not chemically active and are  
274 constant over a distance  $\Delta x$  spanning a significant portion of the city. The next assumption is  
275 that a CH<sub>4</sub> plume measured upwind of the city is well mixed within a layer of depth  $H$  (which is  
276 the same as BLH). We treat wind speed  $u$  as constant within the layer for every hour considered.



277 Given the above-mentioned assumptions we can write a continuity equation describing mass  
278 conservation of CH<sub>4</sub> concentration  $C$  within a box in the following fashion,

$$\Delta x H \frac{\partial C}{\partial t} = \Delta x Q_a + uH(C_b - C) + \Delta x \frac{\partial H}{\partial t} (C_a - C) \quad (3)$$

279 where  $C_b$  is the CH<sub>4</sub> concentration upwind of the city (or background), and  $C_a$  is the CH<sub>4</sub>  
280 concentration above the mixed layer (Hanna et al., 1982; Arya, 1999; Hiller et al., 2014). The  
281 left hand of the equation represents the change in CH<sub>4</sub> concentration with time,  $\Delta x Q_a$  denotes a  
282 constant CH<sub>4</sub> source over the distance  $\Delta x$ ,  $uH(C_b - C)$  indicates a change of CH<sub>4</sub> concentration  
283 due to horizontal advection, and finally  $\Delta x \frac{\partial H}{\partial t} (C_a - C)$  term accounts for the vertical advection  
284 and encroachment processes that result from changing BLH. By assuming steady state  
285 conditions ( $\frac{\partial C}{\partial t} = 0$  and  $\frac{\partial H}{\partial t} = 0$ ), the equation can be simplified to

$$Q_a = \frac{uH(C - C_b)}{\Delta x} \quad (4)$$

286 We use equation 4 to estimate hourly CH<sub>4</sub> emissions ( $Q_a$ ) from Indianapolis (see  
287 assumptions in the paragraph below) given hourly averaged data of  $H$  from the lidar positioned  
288 in the city, wind speed ( $u$ ) from the local weather stations, and upwind ( $C_b$ ) and downwind ( $C$ )  
289 CH<sub>4</sub> mole fractions measured (and then converted to concentrations) at towers 1, 8, and 13  
290 (depending on a wind direction) using data from heights of 40 m, 41 m, and 87 m respectively  
291 (see Fig. 2).

292 The CH<sub>4</sub> concentrations are derived from CH<sub>4</sub> mole fractions by approximating average  
293 molar density of dry air (in mol m<sup>-3</sup>) within the boundary layer for every hour of the day, where  
294 variability of pressure with altitude is calculated using barometric formula and it is assumed that  
295 temperature decreases with altitude by 6.5 K per kilometer. The hourly surface data for pressure  
296 and temperature is taken from KIND weather station. The difference between concentrations

297  $C - C_b$  is instantaneous and not lagged, where  $C_b$  represents air parcel entering the city and  $C$   
298 represents the same air parcel exiting the city (Turnbull et al., 2015). The  $\text{CH}_4$  enhancements  
299  $C - C_b$  are estimated for daytime by averaging observations spanning 12-16 LST and for  
300 nighttime by averaging observations spanning 20-5 LST. These time periods are based on lidar  
301 estimations of when on average  $H$  varies the least. The day and night were required to contain at  
302 least 3 and 9 hourly  $\text{CH}_4$  values respectively for averaging to occur, otherwise the day/night is  
303 eliminated. Observations when  $H$  is below 100 m are not used to avoid the cases when  
304 measurements from towers may be above the boundary layer. In order to better achieve the  
305 assumption that the boundary layer is fully mixed (especially at night), all hours with wind  
306 speeds below 4 m/s are eliminated (Van De Wiel., 2012). To approximate the emissions of the  
307 whole city we need to know the approximate area of the city and the distance over which the  
308 plume is affected by the city  $\text{CH}_4$  sources. The area of the city is about 1024 km<sup>2</sup> (the area of  
309 Marion County) and the length that plume traverses when it is over the city ranges from 32 to 35  
310 km depending on which downwind tower is used. We assume that  $\text{CH}_4$  measurements at towers  
311 8 and 13 are representative of a vertically well-mixed city plume as the towers are located  
312 outside of the city boundaries and allow for sufficient vertical mixing to occur. For S and SW  
313 wind directions tower 8 observations are used to represent downwind conditions with  
314 background observations coming from towers 1 and 13, respectively (based on criterion 1 shown  
315 in Table 1). For W wind direction, tower 13 observations represent the downwind with  
316 background obtained from tower 1. The wind direction is required to be sustained for at least 2  
317 hours, otherwise the data point is eliminated.

318

## 319 **2.7 Indianapolis $\text{CH}_4$ sources**

320 Only a few known CH<sub>4</sub> point sources exist within Indianapolis (Cambaliza et al., 2015, Lamb et  
321 al., 2016). The Southside Landfill (SSLF), located near the center of the city, is thought to be the  
322 largest point source in the city with emissions ranging between about 28 mol/s (inventory from  
323 Maasakkers et al., 2016 and GHG reporting program) and 45 mol/s (aircraft; Cambaliza et al.,  
324 2015) depending on an emission estimation methodology. SSLF could account for as little as  
325 33% (top-down from Cambaliza et al., 2015) or as much as 63% (inventory from Maasakkers et  
326 al., 2016) of total Marion County CH<sub>4</sub> emissions contingent on how much of the total city  
327 emissions are coming from NG. Other city point sources are comparatively small; the  
328 wastewater treatment facility located near SSLF contributes about 3-7 mol/s (inventory from  
329 Lamb et al. 2016), and the transmission-distribution transfer station at Panhandle Eastern  
330 Pipeline (also known as a city gate and further in this study abbreviated as PEP) is estimated to  
331 be about 1 mol/s (inventory from Lamb et al. 2016). The remaining CH<sub>4</sub> sources, mainly from  
332 NG and livestock, are considered to be diffuse sources and are not well known. Potential sources  
333 of emissions related to NG activities include gas regulation meters, emissions from transmission  
334 and storage, and Compressed Natural Gas (CNG) fleets. These diffuse NG sources account for  
335 21-67% (this value varies due to the uncertainty in SSLF emissions) of the city emissions or 20  
336 mol/s (inventory from Maasakkers et al., 2016) to 64 mol/s (top down from Cambaliza et al.,  
337 2015). Livestock emissions for Marion County are estimated to be around 1.5 mol/s (inventory  
338 from Maasakkers et al., 2016). An important question remains of whether SSLF or NG is the  
339 dominant CH<sub>4</sub> source in Indianapolis. There could also be a possibility of temporal variability in  
340 either of the sources as described in the section above.

341

## 342 **3 Results and discussion**

343

### 344 **3.1 Inversion and city boundaries**

345 A significant portion of CH<sub>4</sub> emissions across the U.S. can be characterized by numerous large  
346 point sources scattered throughout the country rather than by broad areas of smaller  
347 enhancements (Maasakkers et al., 2016). Because of this, the total emissions for a given domain  
348 can be very sensitive to how that domain is defined. A small increase or decrease in the domain  
349 area could add or remove a large point source and significantly impact the total emissions  
350 defined within the domain.

351 In the case of Indianapolis, this issue became apparent when the emissions were  
352 calculated using an atmospheric inversion model (Lamb et al., 2016; Lauvaux et al., 2016). The  
353 atmospheric inversion solved for fluxes in domain 1 (Fig. 2), which significantly increased the  
354 estimated emissions in comparison with the inventory values that were gathered mainly within  
355 Marion County (domain 2). When reduced to domain 2, inverse modeling emission estimates  
356 decrease to 107 mol/s, which falls within an error bar of Lamb et al. (2016) inventory estimate.  
357 This difference is significant and could at least partially explain the discrepancy shown in Figure  
358 1 between the emission values from the inventories and emission results from the inverse  
359 modeling. However, even the decreased inverse modeling estimate is about 40% higher than the  
360 inventories.

361 It is difficult for us to critically assess this result without performing complex sensitivity  
362 analysis with this inversion system (which is not the goal of this article), but it is important to  
363 note that inverse modeling is prone to errors in prior, background, and meteorological transport.  
364 Also, Figure 3 indicates that tower data were sparse in 2012-2013 perhaps contributing to  
365 potential error in the inversion.

366 Additionally, the subject of the domain is relevant for airborne mass balance flights  
367 because a priori the magnitude and variability of background plume is unknown and could be

368 easily influenced by upwind sources. The issue of background is discussed further in the next  
369 section.

370

### 371 **3.2 Variability in CH<sub>4</sub> background**

372 Comparisons between criterion 1 and criterion 2 CH<sub>4</sub> background mole fractions as a  
373 function of wind speed and direction are visualized using frequency and bivariate polar plots  
374 (Fig. 4). Both backgrounds generally agree on the higher CH<sub>4</sub> originating from the SW, SE, and  
375 E wind directions (Figs. 4c-f); however, the values themselves differ especially when winds are  
376 from NW, SW, and SE. As background difference plots (Figs. 4g-h) indicate, there is a  
377 noticeable variability between the magnitudes of the CH<sub>4</sub> backgrounds, where criterion 2, by  
378 design, typically has higher background mole fractions. The background differences, at a given  
379 hour, suggest that the CH<sub>4</sub> field enveloping the city is heterogeneous with differences between  
380 towers ranging from 0 to over 45 ppb (Fig. 4g). Because large gradients in CH<sub>4</sub> background over  
381 the city could pose challenges for flux estimations using top down methods such as inverse  
382 modeling and aircraft mass balance, it is imperative to establish whether the background  
383 differences vary randomly or systematically and how to choose a background to minimize these  
384 errors.

385 To further understand the nature of background variability we calculate the mean,  
386 standard deviation, and standard error of background hourly differences between criterion 2 and  
387 criterion 1 from November 2014 to December 2016 for each of the eight wind directions  
388 mentioned in Table 1. The results are shown in Figure 5. Systematic bias is evident for the SE,  
389 S, SW, W, and NW wind sectors, whereas random error dominates N, NE, and E wind  
390 directions. Wind directions showing statistically significant bias have mean biases ranging from

391 2 to 5 ppb, with values as large as 8 ppb falling within the range of  $2 \times$  standard error. Standard  
392 deviation plot indicates potential background discrepancy that can occur on any given day, where  
393 W wind direction is the least variable with  $2 \times$  standard deviation close to 20 ppb, while SE wind  
394 direction is the most variable with  $2 \times$  standard deviation falling at about 50 ppb.

395 Random errors in the mole fractions of background differences (biases) are also  
396 important and are a function of the length of the data record. We quantify the random error in  
397 the CH<sub>4</sub> background mole fraction differences using the bootstrap method by randomly sampling  
398 2 to 150 hours (small and large sample size) of the background CH<sub>4</sub> differences for each of the  
399 wind directions with replacement (we make the assumption that our differences are independent  
400 since we eliminated lag 1 autocorrelation from the data). This sub-sampling experiment is  
401 repeated 5000 times (Efron and Tibshirani, 1986). The standard deviations of the mean  
402 (standard error) of the 5000 simulated differences are calculated for each wind direction. The  
403 resulting standard errors of the city CH<sub>4</sub> background differences, multiplied by 2 to represent the  
404 95% confidence intervals, are shown as a function of the length of the data record in Figure 6.  
405 Because random error falls as sample size grows it makes sense to assign a threshold indicating a  
406 minimum number of samples needed to achieve a theoretical precision for each wind direction.

407 One way to assign a required precision would be to make sure that the standard error  
408 (random error) reaches a point where it is less than Indianapolis enhancement of about 12 ppb (a  
409 higher estimate of the Indianapolis enhancement from section 3.3) by a factor of 2 when  
410 combined with a bias (Table 2). Meaning that the sum of bias and standard error must be at most  
411 6 ppb. In this approach each wind direction would have a different threshold because of the  
412 differences in biases. For instance, given this requirement NW direction would need a random  
413 error of 1 since its bias is 5. For NW direction, this threshold would require more than 150

414 samples. For N direction on the other hand, where the bias is 1, the requirement is fulfilled when  
415 random error crosses 5 ppb at 74 samples. Now we consider these random and systematic errors  
416 in CH<sub>4</sub> background differences in the context of Indianapolis urban CH<sub>4</sub> emissions.

417 For Indianapolis, using INFLUX tower network, we estimated that depending on sample  
418 size (number of hours sampled) and wind direction, background gradient across the city over 12-  
419 16 LST could vary from 0 to about 50 ppb (Fig. 5b). Given that the average afternoon CH<sub>4</sub>  
420 enhancement of the city is around 8-12 ppb (section 3.3; Fig. 7; Cambaliza et al., 2015; Miles et  
421 al., 2017), the error on the estimated emissions could easily be over 100% if the analysis does not  
422 approach the issue of background with enough sampling. A sample size of about 50 independent  
423 hours significantly decreases background uncertainty for N, NE, E, S, and W wind directions and  
424 allows for a more accurate assessment of the CH<sub>4</sub> emissions at Indianapolis. For CH<sub>4</sub> sources  
425 with a significantly larger signal than their regional background, the mentioned background  
426 variability becomes less impactful on results, but because Indianapolis is a relatively small  
427 emitter of CH<sub>4</sub>, and because there are relatively large sources outside of the city, uncertainties  
428 due to background estimation are comparatively large. Our uncertainty assessment suggests that  
429 the highly variable CH<sub>4</sub> emission values of Indianapolis from aircraft mass balance calculations  
430 shown in Figure 1 are at least partially due to the variability in the urban CH<sub>4</sub> background of  
431 Indianapolis.

432

### 433 **3.3. Temporal variability of methane enhancements and fluxes in Indianapolis**

434 Figure 7 presents average CH<sub>4</sub> mole fraction enhancements and flux calculations  
435 (equation 4) at towers 8 and 13 for years 2014, 2016, and 2013-2016 (for the detailed  
436 methodology see section 2.6). The years of 2014 and 2016 are chosen for temporal comparison

437 because they do not contain major BLH data gaps. The error bars in the figure show the standard  
438 error multiplied by 2 indicating 95% confidence interval of each average.

439 One of the more interesting features in the Figure 7 is a day/night variability of CH<sub>4</sub>  
440 emissions at Indianapolis. The most prominent example of this feature is found in Figure 7c,  
441 where the estimates for both years suggest that daytime emissions are approximately twice as  
442 large as the emissions at night. The decrease of the CH<sub>4</sub> emissions at night also appears in tower  
443 13, but the errors are too high in those estimates to make any definitive conclusions. A similar  
444 urban CH<sub>4</sub> emissions diurnal variability is reported by Helfter et al. (2016) in their study of  
445 GHGs for London, UK, where they attribute diurnal variation of CH<sub>4</sub> emissions to the NG  
446 distribution network activities, fugitive emissions from NG appliances, and to temperature-  
447 sensitive CH<sub>4</sub> emission sources of biogenic origin (such as a landfill). Taylor et al. (2018)  
448 suggest that CH<sub>4</sub> emissions from landfills exhibit a diurnal cycle with higher emissions in early  
449 afternoon and 30-40% lower emissions at night.

450 With regard to yearly temporal variability we are only able to compare years 2014 and  
451 2016 due to limited BLH data for other years. Results from both towers suggest that  
452 Indianapolis overall CH<sub>4</sub> emissions did not change significantly between 2014 and 2016.  
453 Although it is important to be cautious about interpreting actual flux estimations given the  
454 assumptions mentioned in section 2.6, it is interesting to note that the flux values from both  
455 towers average at about 70 mol/s, which puts our value right in between inventory and inversion  
456 estimates shown in Figure 1. If we assume that SSLF emissions are generally known (GHG  
457 reporting program) that would indicate that emissions from NG distribution are likely to be  
458 somewhat higher than both of the inventories currently estimate and consistent with higher error  
459 bar of Lamb et al. (2016) calculation. Another possible scenario is that SSLF emissions are



460 higher than what is currently assumed. Given these complexities, uncertainty regarding the exact  
461 emissions from NG distribution at Indianapolis still remains.

462

### 463 **3.4 Methane Sources in Indianapolis**

464 Bottom-up emission inventories have difficulty tracking changes in sources over time. Our  
465 continuous tower network observations can monitor temporal and spatial variability in sources of  
466 CH<sub>4</sub> in Indianapolis. To do so we employ the aforementioned bivariate polar plots to verify  
467 known sources and potentially identify unknown sources across the city. We compare two time  
468 periods, 2014-2015 (two full years) and 2016. Figure 8 displays bivariate polar plots of CH<sub>4</sub>  
469 enhancements using criterion 1 background at 9 INFLUX towers in Indianapolis over the two  
470 years of 2014 and 2015. Figure 9 shows the same plot, but for the year 2016. Here we have  
471 separated 2016 from 2014-2015 because of different results noted during this time.

472 The images reveal that the most consistent and strongest source in the city is the SSLF.  
473 This is most evident from the 40+ ppb CH<sub>4</sub> enhancements detected at towers 7, 10 and 11  
474 coming from the location of the SSLF (by triangulation). Enhancements from the landfill appear  
475 to also be detectable at towers 2, 4, 5, and 13. Based on these observations it can be concluded  
476 that there is no other source in Marion County comparable in strength to the SSLF. A small  
477 fraction of the SSLF plume is likely due to the co-located wastewater facility, but the inventory  
478 estimates suggest that the wastewater treatment facility is responsible for no more than 7% of  
479 this plume (Cambaliza et al., 2015; Massackers et al., 2016). The PEP, located in the  
480 northwestern section of the city, may be partially responsible for a plume of 5-10 ppb at towers 5  
481 and 11. However, the plume is less detectable using the criterion 2 background value that has  
482 higher background (using tower 8 as a background) from NW wind direction (not shown),

483 adding uncertainty to the true magnitude of the enhancement from this source. The same is true  
484 for towers 2 and 13, which have pronounced plumes when winds are from the NW with the  
485 criterion 1 background, but when background 2 is used these plumes vanish (not shown). Such  
486 inconsistency makes it difficult to attribute these plumes to an urban source.

487 Another important point is the cluster of large enhancements surrounding tower 10 in  
488 2014 - 2015. Because no other tower sees these enhancements (at least at comparable  
489 magnitudes), we believe that these plumes are the result of local NG leaks likely from residential  
490 sector of Indianapolis. These plumes are not consistent temporally or spatially as they mostly  
491 disappear in 2016, potentially indicating that they are transient and localized NG distribution  
492 leaks. It is difficult to ascertain the exact combined magnitude of these leaks since they mix  
493 together with SSLF into an aggregated city plume when observed from downwind towers such as  
494 8 and 13. Yet, none of these leaks appear to be even remotely close in magnitude to the  
495 emissions that originate from SSLF. Thus, the diffuse NG source suspected to be twice as large  
496 as the SSLF source (Lamb et al., 2016) does not appear to be supported by these data. This  
497 assertion questions conclusions made by Cambaliza et al., (2015), who attributed most of the  
498 CH<sub>4</sub> emitted by Indianapolis to NG related activities. We hypothesize that the relatively high  
499 Indianapolis CH<sub>4</sub> emissions (see Fig. 1) reported by Cambaliza et al. (2015) are the result of the  
500 low sample size of airborne flux estimates making it prone to large random errors (see section  
501 3.2). However, it is imperative to be careful and acknowledge the limitations of the current  
502 analysis. Our flux estimations at towers 8 and 13 discussed in the previous section do imply that  
503 emissions from NG distribution may be higher than estimated by the inventories indicating that  
504 an overall NG contribution may be comparable in strength to SSLF. This discrepancy requires  
505 further investigation.

506

#### 507 **4 Conclusions**

508 We have examined four specific contributions to discrepancies between urban top-down and  
509 bottom-up CH<sub>4</sub> emission estimates from Indianapolis: domain definition, heterogeneous  
510 background mole fractions, temporal variability in emissions, and source knowledge. Results  
511 indicate that the urban domain definition is crucial for the comparison of the emission estimates  
512 among various methods. Atmospheric inverse flux estimates for Marion County, which is  
513 similar to the domain that is analyzed by inventory and airborne mass balance methodologies  
514 (Mays et al., 2009, Cambaliza et al., 2014, Lamb et al., 2016), is 107 mol/s compared to 160  
515 mol/s that is estimated for the larger domain (Hestia inventory domain; Gurney et al., 2012).  
516 This partially explains higher emissions in inverse modeling estimates shown by Lamb et al.  
517 (2016); however, 107 mol/s is still about 40-50% higher than what EPA and Lamb et al. (2016)  
518 find in their inventories (Fig. 1). Although it is difficult to generalize with certainty regarding  
519 this particular inversion, significant errors are possible in an inversion system due to lack of data,  
520 false assumptions regarding prior, biased background, and erroneous modeled meteorological  
521 transport.

522 To better understand background variability at Indianapolis two different but acceptable  
523 background towers are selected based on specific criteria for each wind direction and their  
524 differences are used to assess heterogeneity of CH<sub>4</sub> background at Indianapolis. Background  
525 criterion 1 looks for a tower that is consistently lower than other towers, while background  
526 criterion 2 picks a tower that is outside of Marion County domain and is not downwind of any  
527 nearby sources as determined by EPA 2012 inventory. We focus on midday atmospheric  
528 conditions to avoid the complexities of vertical stratification in the stable boundary layer. The

529 midday Indianapolis atmospheric CH<sub>4</sub> mole fraction background is shown to be heterogeneous  
530 with 2-5 ppb statistically significant biases for NW, W, SW, S and SE wind directions. Random  
531 errors of background differences are a function of sample size and decrease as a number of  
532 independent samples increase. Low sample volumes, such as a few hours of data from a single  
533 point, are prone to random errors on the order of 10-30 ppb in the CH<sub>4</sub> background, similar to the  
534 magnitude of the total enhancement from the city of Indianapolis, which is estimated to be on  
535 average around 10-12 ppb. Longer-term sampling and/or more extensive background sampling  
536 are necessary to reduce the random errors. Sample size required to reduce random errors of  
537 background differences to an acceptable value for flux calculation is largely dependent on a wind  
538 direction. Both bias (long-term average of background differences) and its random error are  
539 important when estimating total background uncertainty. The results indicate that N, NE, E, S,  
540 and W wind directions are more favorable for flux estimation and would require multiple days of  
541 measurements (e.g. about 50 independent hours of measurements) to reduce background  
542 uncertainty to about 6 ppb, noticeably smaller than the typical CH<sub>4</sub> enhancement from  
543 Indianapolis. The remaining wind directions would require over 150 independent hourly  
544 measurements to achieve similar precision. We also estimate that depending on a wind direction  
545 for any given hour the spatial variability in background can be anywhere from 0 to 50 ppb. This  
546 uncertainty in the CH<sub>4</sub> background may partially explain Heimburger et al. (2017) finding of  
547 large variability in airborne estimates of Indianapolis CH<sub>4</sub> emissions. Given many samples, the  
548 airborne studies converge to an average value of CH<sub>4</sub> flux that is noticeably closer to the  
549 inventory estimates for Indianapolis than their individual components as presented in Figure 1.

550 Measurement and analysis strategies can minimize the impacts of these sources of error.  
551 Spatially extensive measurement of upwind CH<sub>4</sub> mole fractions are recommended. For towers or

552 other point-based measurements, multiple upwind measurement locations are clearly beneficial.  
553 For the aircraft mass balance approach, we recommend an upwind transect to be measured,  
554 lagged in time if possible, to provide a more complete understanding of the urban background  
555 conditions. Complex background conditions might suggest that data from certain days or wind  
556 directions should not be used for flux calculation. Finally, a mesoscale atmospheric modeling  
557 system informed with the locations of important upwind CH<sub>4</sub> sources can serve as a powerful  
558 complement to the atmospheric data (Barkley et al., 2017). Such simulations can guide sampling  
559 strategies, and aid in interpretation of data collected with moderately complex background  
560 conditions.

561 With regard to temporal variability, no statistically detectable changes in the emission  
562 rates were observed when comparing 2014 and 2016 CH<sub>4</sub> emissions. However, a large  
563 difference between day and night CH<sub>4</sub> emissions was implied from a simple budget estimate.  
564 Night (20-5 LST) emissions may be 2 times lower than the emissions during the afternoon (12-  
565 16 LST) hours. Because prior estimates of top-down citywide emissions are derived using  
566 afternoon-only measurements, overall emissions of Indianapolis may be lower than these studies  
567 suggest. This bias may be present in studies performed in other cities as well. Our study  
568 suggests that day/night differences in CH<sub>4</sub> emissions must be understood if regional emission  
569 estimates are to be calculated correctly. Long-term, tower-based observations are an effective  
570 tool for understanding and quantifying multi-year variability in urban emissions.

571 One final point addressed in this study is the location of major CH<sub>4</sub> sources in  
572 Indianapolis. Analysis of the INFLUX observation data suggests that inventories for  
573 Indianapolis are mostly accurate and that there is no clear evidence of a large, diffuse NG source  
574 of CH<sub>4</sub> as implied by Lamb et al. (2016). The only major source in the city is SSLF and it is

575 observed at multiple towers. There is an evidence for occasional NG leaks, but they appear  
576 localized and limited in their strength. However, we cannot completely rule out occasional  
577 significant leaks of CH<sub>4</sub> from NG at Indianapolis due to the nature of our assumptions.

578 Overall, assessment of the CH<sub>4</sub> emissions at Indianapolis highlights a number of  
579 uncertainties that need to be considered in any serious evaluation of urban CH<sub>4</sub> emissions. These  
580 uncertainties amplify for Indianapolis since its CH<sub>4</sub> emissions are comparable in magnitude to  
581 the regional background flow and as our results show it may be difficult at times to distinguish  
582 noise in the background from the actual city emissions signal. The evaluation of larger CH<sub>4</sub>  
583 sources may be easier with respect to separating signal from background. However, all of the  
584 points raised in this work will be nonetheless relevant and need to be addressed for our  
585 understanding of urban CH<sub>4</sub> emissions to significantly improve.

586

### 587 **Author Contribution**

588 Nikolay Balashov, Kenneth Davis, and Natasha Miles developed the study and worked together  
589 on generating the main hypothesis of this work. They also wrote most of the manuscript.  
590 Nikolay Balashov wrote all of the codes and performed the analyses presented in this work as  
591 well as generated all of the figures. Natasha Miles and Scott Richardson helped with  
592 maintenance and gathering of the INFLUX tower data. They also wrote section 2.2 of the paper.  
593 Thomas Lauvaux helped with the analysis presented in Fig. 1 and section 3.1 concerning  
594 interpretation of the inversion modeling results from Lamb et al. (2016). Thomas Lauvaux also  
595 helped with repeating the inversion experiment for two different Indianapolis domains (Fig. 1).  
596 Zachary Barkley significantly contributed to discussions regarding the hypothesis and careful  
597 presentation of sections 2.6 and 3.3. Timothy Bonin provided all of the lidar data and wrote the

598 second part of section 2.3 regarding the lidar and the methodology used to determine planetary  
599 boundary layer heights. He also contributed to sections 2.6 and 3.3.

600

#### 601 **Competing Interests**

602 The authors declare that they have no conflict of interest.

603

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612

613

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856 **Tables**

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858 **Table 1.** INFLUX towers used to estimate CH<sub>4</sub> background based on two different criteria. Numbers in  
 859 bold indicate towers chosen to generate a background field when multiple options are possible (for more  
 860 details see discussion). In short, criterion 1 uses towers with the lowest mean CH<sub>4</sub> for a specific wind  
 861 direction, and criterion 2 uses towers outside of Marion County and not downwind of large sources  
 862 (including the city as a whole).

Wind Direction	CH <sub>4</sub> Background Towers	
	Criterion 1	Criterion 2
North (N)	8	<b>13</b> , 8
Northeast (NE)	8	<b>13</b> , 8, 2
East (E)	<b>2</b> , 8	<b>8</b> , 4, 1, 2
Southeast (SE)	1	<b>8</b> , 13, 4, 1
South (S)	1	<b>4</b> , 13, 1
Southwest (SW)	13	<b>1</b> , 4
West (W)	1	<b>4</b> , 1
Northwest (NW)	1	<b>8</b> , 1

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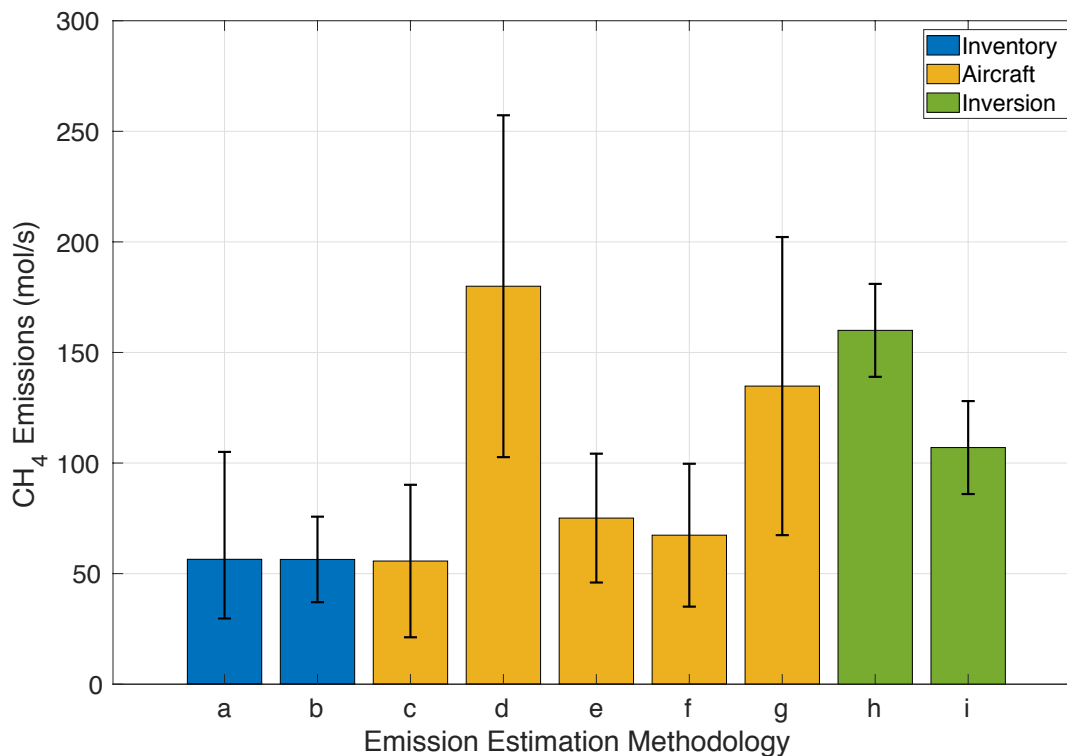
880 **Table 2.** A number of independent samples needed (column 4) to satisfy combined requirement of 6 ppb  
881 background error based on the sum of bias and random error (explained in section 3.2) as a function of  
882 wind direction.

Wind Direction	Bias (ppb)	Threshold (ppb)	Samples Needed
N	1	5	74
NE	1	5	36
E	0.5	5.5	46
SE	4	2	>150
S	2	4	53
SW	4.5	1.5	>150
W	3	3	52
NW	5	1	>150

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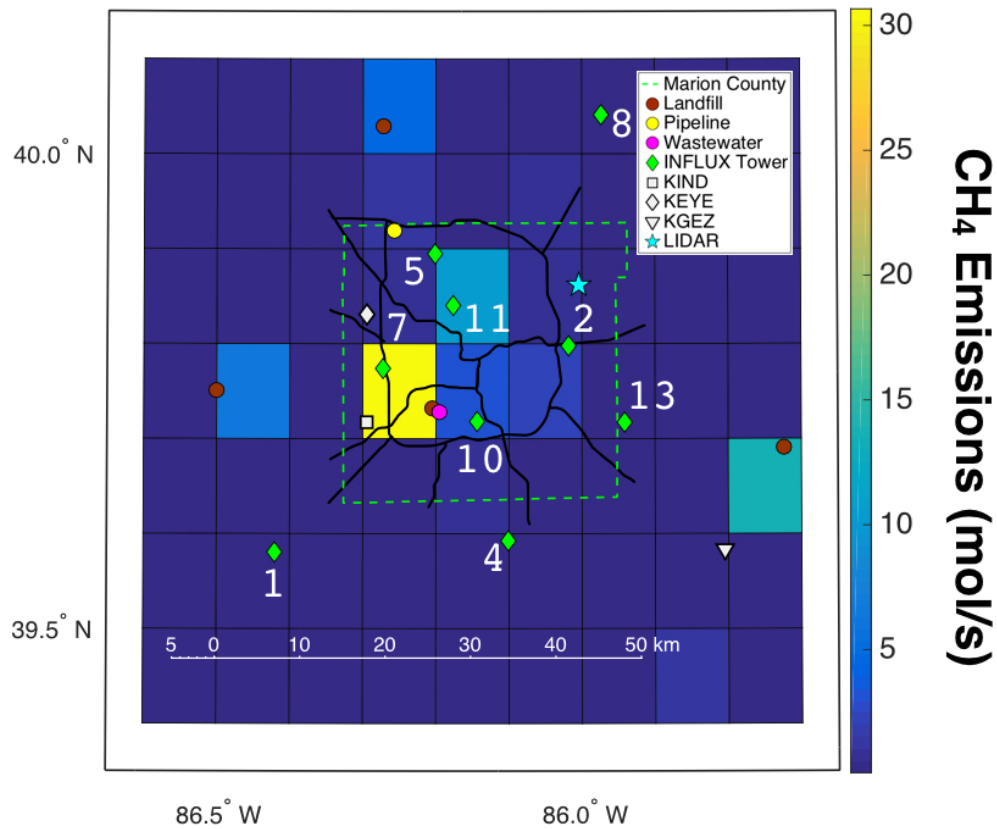
915 **Figures**  
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918 **Figure 1.** Various estimates of CH<sub>4</sub> emissions at Indianapolis. **(a, b)** Bottom-up estimates of CH<sub>4</sub>  
 919 emissions conducted by Lamb et al. (2016) in 2013 and Maasakkers et al. (2016) based on the EPA 2012  
 920 inventory respectively. Error bars show 95% confidence intervals (for more details see above-mentioned  
 921 articles). **(c-g)** Top-down evaluations of CH<sub>4</sub> emissions with aircraft from various flight campaigns where  
 922 **(c)** contains 5 flights over March-April of 2008, **(d)** contains 3 flights over November-January of 2008-  
 923 09, **(e)** contains 5 flights over April-July of 2011, **(f)** contains 9 flights from November-December, 2014,  
 924 and **(g)** contains the same 5 flights over April-July of 2011 as in (e) but uses different methodology.  
 925 Methodologies for **(c-f)** are described in Lamb et al. (2016) and methodology for **(g)** is described in  
 926 Cambaliza et al. (2015). Error bars show 95% confidence intervals (for more details see above-  
 927 mentioned articles). **(h, i)** Top-down evaluations of CH<sub>4</sub> emissions for 2012-2013 using tower inversion  
 928 modeling methodology with two different domains, where **(h)** uses the full domain of Figure 2 and **(i)**  
 929 uses only the Marion County domain of Figure 2. The inversion methodology and 95% confidence  
 930 intervals are described in detail in Lamb et al. (2016).

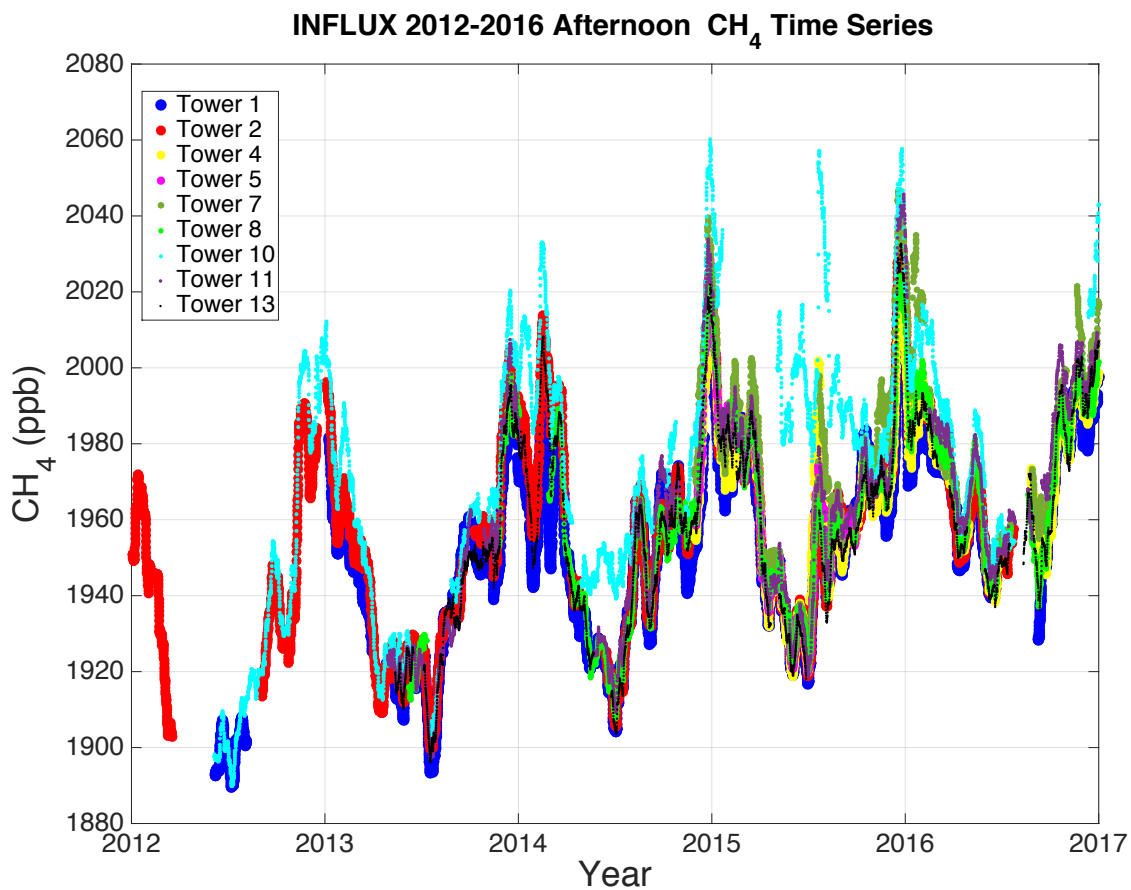
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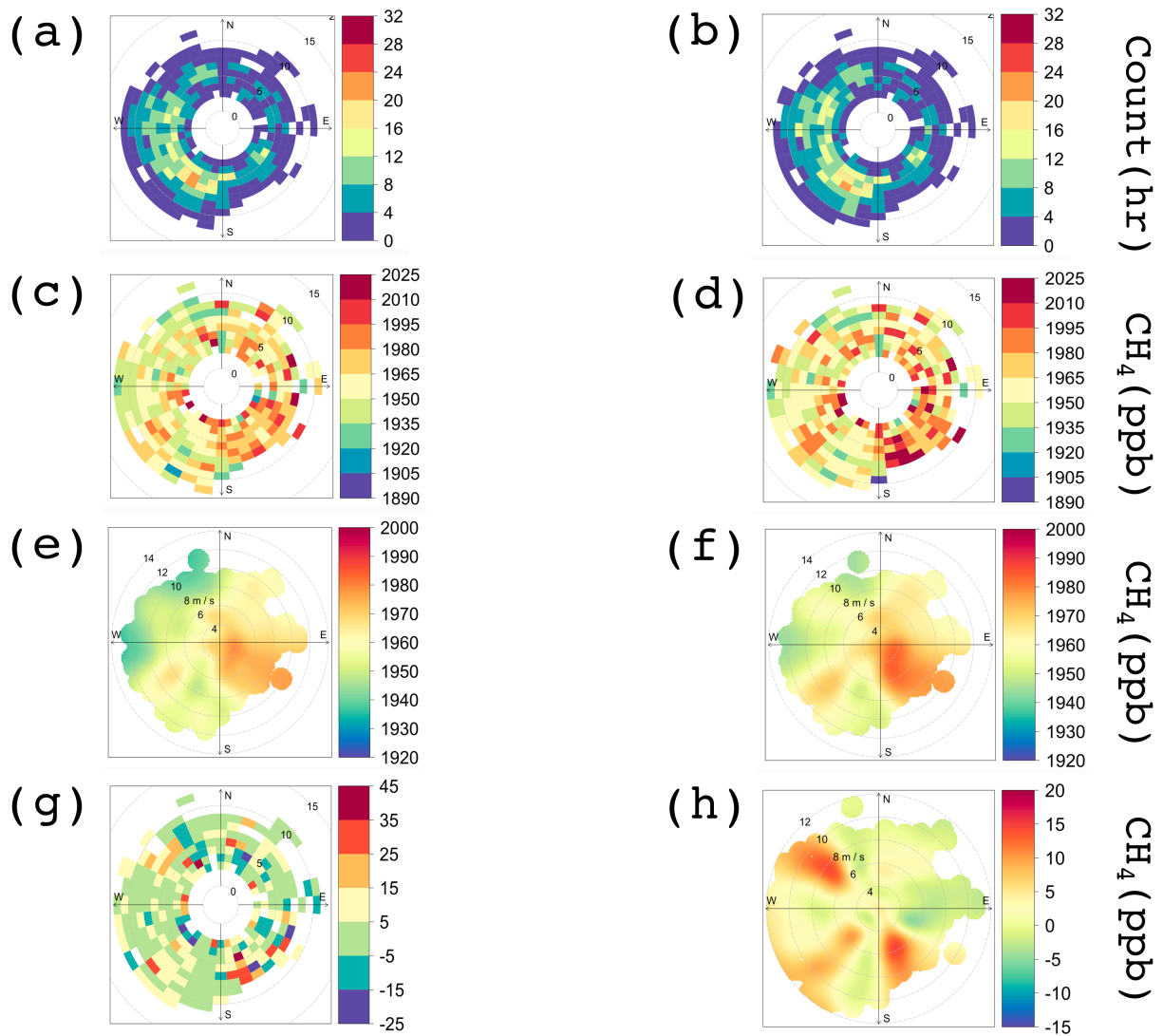
936 **Figure 2.** Map of the primary roads in Indianapolis, INFLUX towers, lidar system, weather stations, and  
 937 a few CH<sub>4</sub> point sources plotted over the gridded CH<sub>4</sub> emissions (mol/s) from the EPA 2012 Inventory  
 938 (Maasakkers et al., 2016). The gridded map of emissions includes emissions from the mentioned point  
 939 sources; their position is provided to aid in interpretation of the observations. The dashed bright green  
 940 line denotes Marion County borders.

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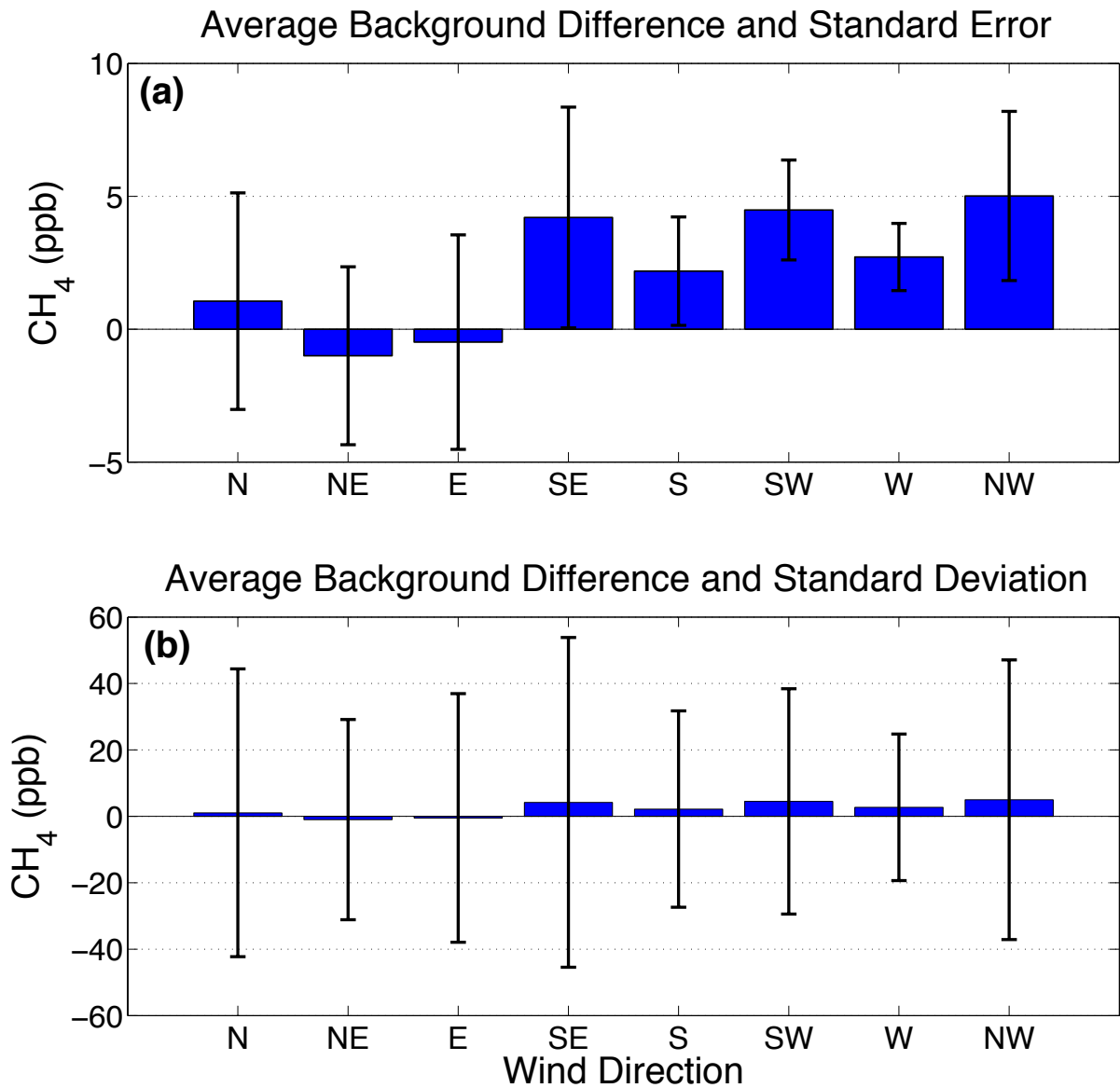
943 **Figure 3.** 20-day running average of afternoon (12-16 LST; the hours are inclusive) CH<sub>4</sub> mole fractions  
 944 as measured by the INFLUX tower network (highest available height is used) from 2012 through 2016.



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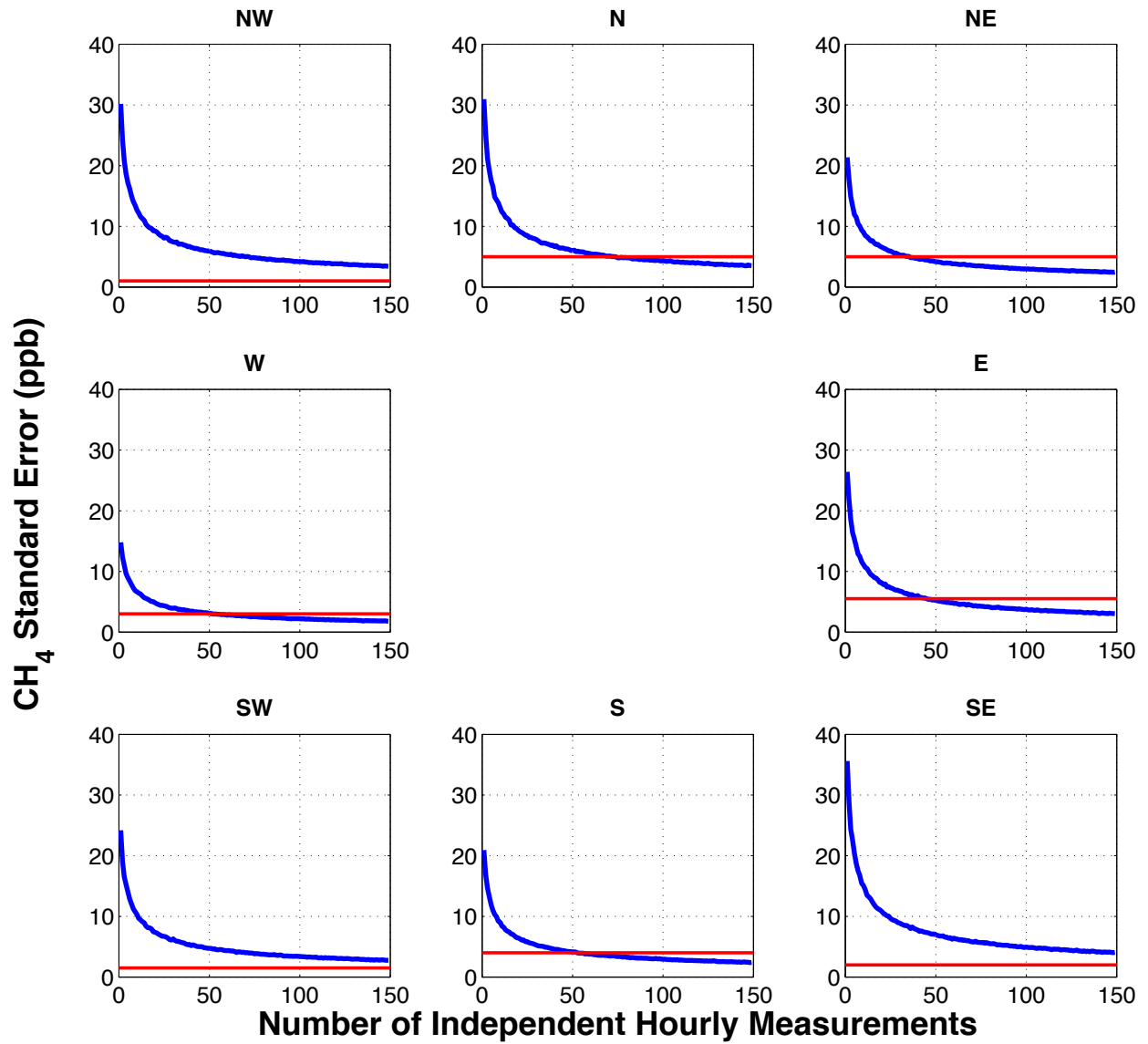
946 **Figure 4.** Frequency and bivariate polar plots of  $\text{CH}_4$  background for Indianapolis using data from 12-16  
 947 LST, November 2014 through December 2016 given 2 different criteria (Table 1). **(a)** Polar histogram  
 948 indicating a number of hourly measurements available using criterion 1. **(b)** Same as (a) only for criterion  
 949 2. Differences between (a) and (b) are due to slight differences in data availability at the considered  
 950 towers. **(c)** Polar frequency plot of the  $\text{CH}_4$  background using criterion 1. **(d)** Same as (c) only for  
 951 criterion 2. **(e)** Polar bivariate plot of  $\text{CH}_4$  background using criterion 1. **(f)** Same as (e) only for  
 952 criterion 2. **(g)** Polar frequency plot of difference between the backgrounds: *criterion 2* – *criterion 1*. **(h)** Same  
 953 as (g) but shown with a bivariate polar plot.

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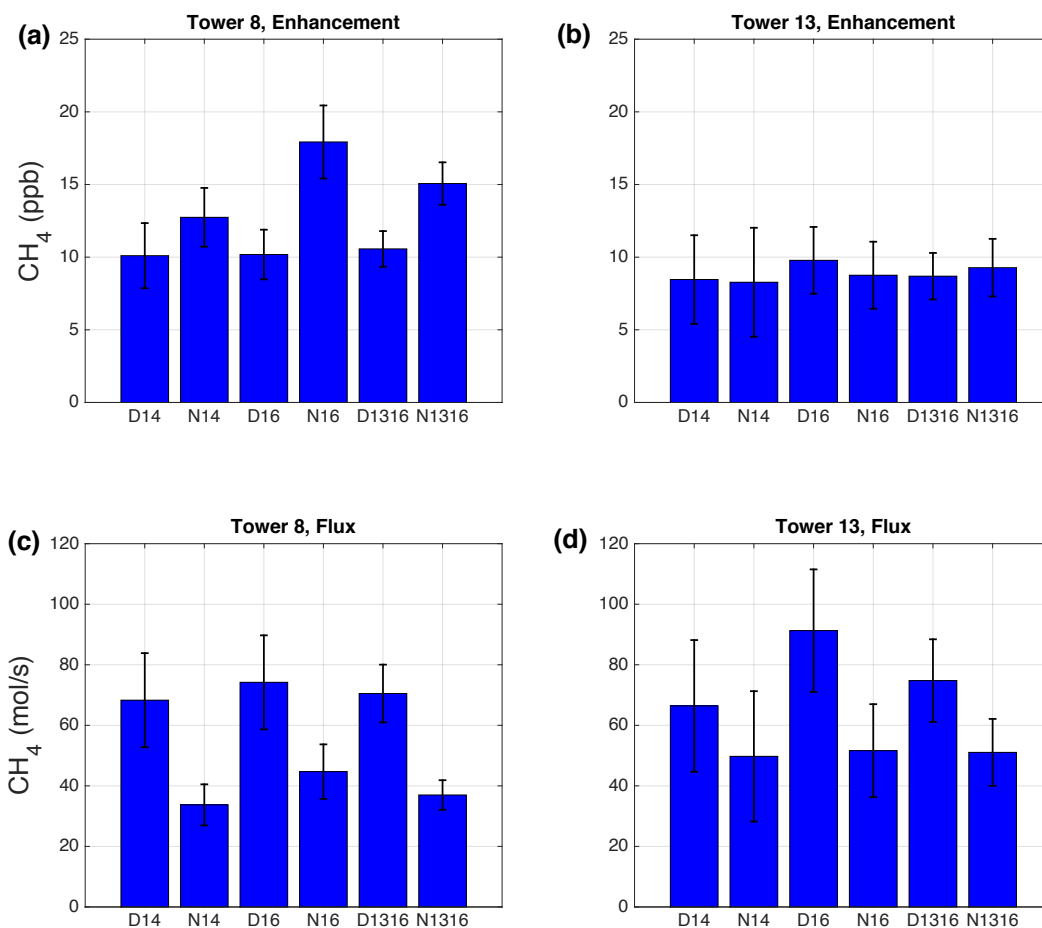


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 956 **Figure 5.** Average of the differences between criteria 2 and 1 CH<sub>4</sub> backgrounds at Indianapolis as a  
 957 function of wind direction. These averages are generated from the same data that is used in Figure 4 and  
 958 reflect results shown in Figure 4g. Error bars indicate in (a) 2 × standard error and in (b) 2 × standard  
 959 deviation.

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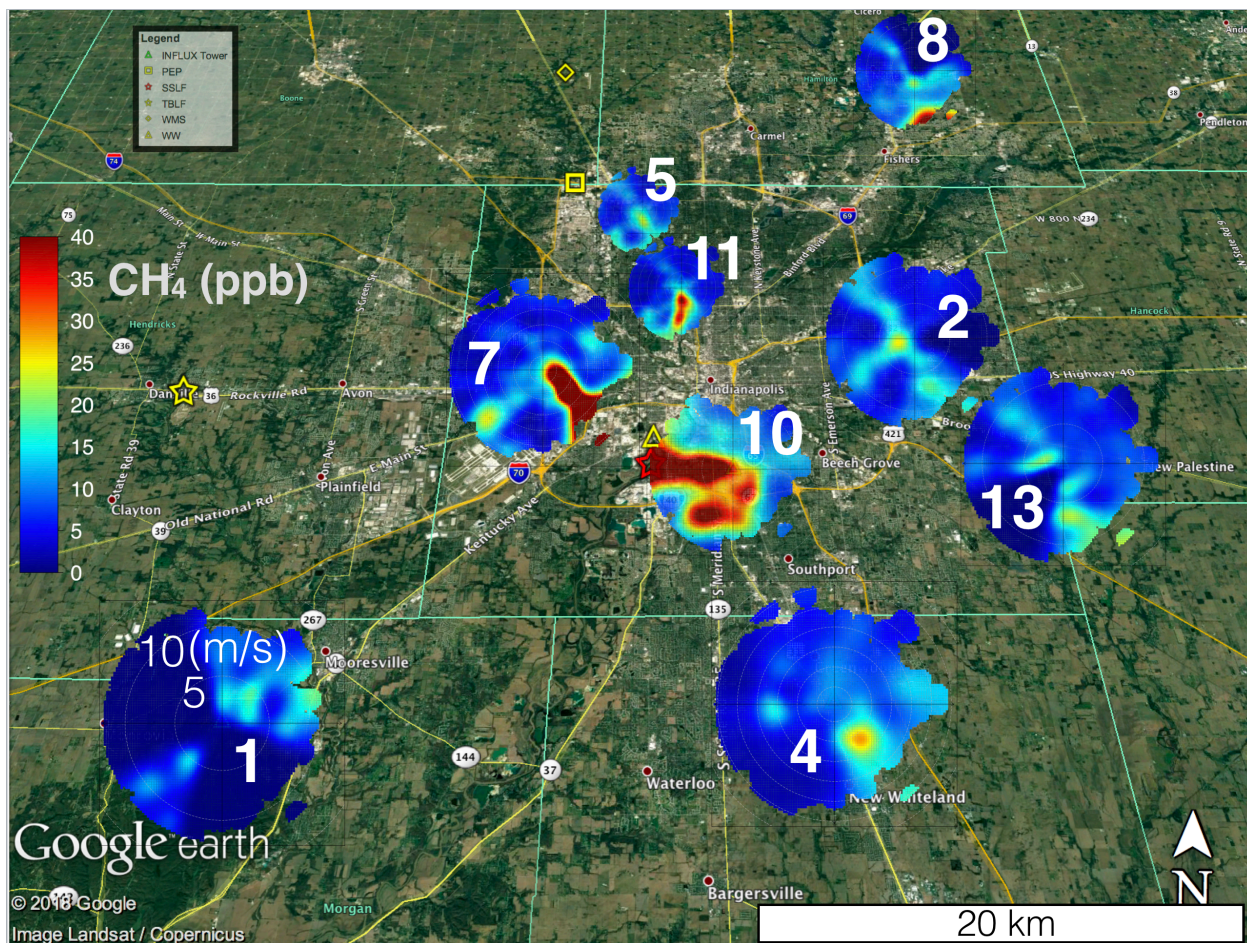


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 962 **Figure 6.** Bootstrap simulation of the standard errors  $\times 2$  in Indianapolis CH<sub>4</sub> background mole fraction  
 963 differences (between criteria 2 and 1) as a function of sample size and wind direction (see text for details).  
 964 Thresholds for each of the wind directions indicate a random error threshold needed for the background  
 965 uncertainty to be within 50% of Indianapolis CH<sub>4</sub> enhancement of 12 ppb.



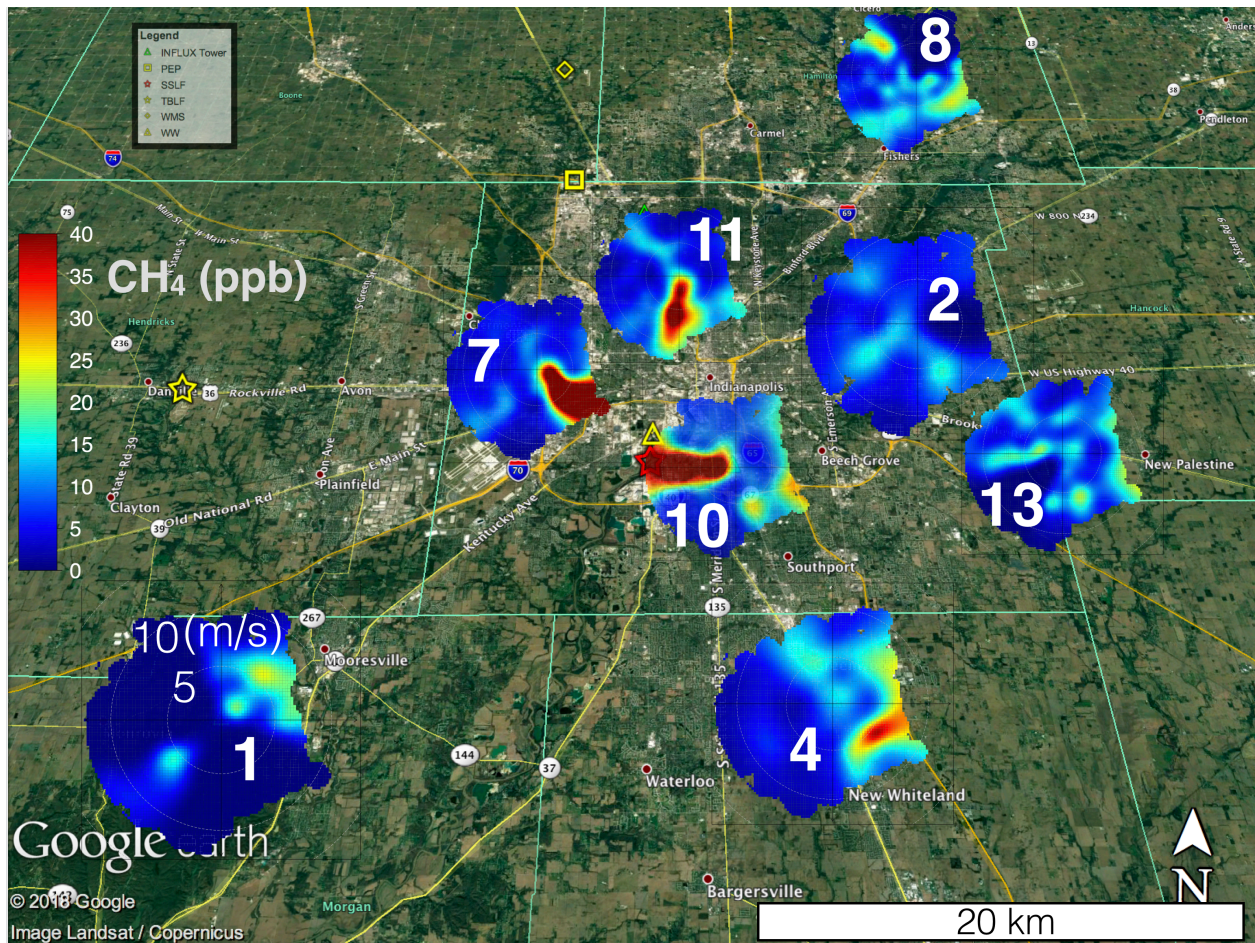
968 **Figure 5.** Averages of the daytime (D) and nighttime (N) CH<sub>4</sub> enhancements and fluxes at INFLUX  
 969 towers 8 and 13 for years 2014 (14), 2016 (16), and 2013-2016 (1316). The error bars represent 95%  
 970 confidence interval of each mean value. **(a)** Estimates of CH<sub>4</sub> enhancements from tower 8. **(b)** Estimates  
 971 of CH<sub>4</sub> enhancements from tower 13. **(c)** Estimates of CH<sub>4</sub> flux from tower 8. **(d)** Estimates of CH<sub>4</sub> flux  
 972 from tower 13.





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 974 **Figure 6.** Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH<sub>4</sub> enhancements  
 975 at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for full years of 2014 and  
 976 2015 over the afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites follow  
 977 the same convention. Legend indicates known sources of CH<sub>4</sub>: Panhandle Eastern Pipeline (PEP),  
 978 Southern Side Landfill (SSLF), Twin Bridges Landfill (TBLF), Waste Management Solutions (WMS),  
 979 and Waste Water treatment facility (WW). The known magnitudes of sources that are in Marion County  
 980 (PEP, SSLF, and WW) are reported in section 2.7. Magnitudes of TBLF and WMS according to EPA are  
 981 approximately 5 mol/s. The largest known source on the map is SSLF.





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983 **Figure 7.** Google Earth image overlaid with bivariate polar plots (section 2.5) of the CH<sub>4</sub> enhancements  
 984 at 9 INFLUX towers in Indianapolis using the criterion 1 background (Table 1) for year 2016 over the  
 985 afternoon (12-16 LST). The wind speed scale is only labeled at site 1; other sites follow the same  
 986 convention. Legend indicates known sources of CH<sub>4</sub>: Panhandle Eastern Pipeline (PEP), Southern Side  
 987 Landfill (SSLF), Twin Bridges Landfill (TBLF), Waste Management Solutions (WMS), and Waste Water  
 988 treatment facility (WW). The known magnitudes of sources that are in Marion County (PEP, SSLF, and  
 989 WW) are reported in section 2.7. Magnitudes of TBLF and WMS according to EPA are approximately 5  
 990 mol/s. The largest known source on the map is SSLF.