



## Source decomposition of eddy-covariance CO<sub>2</sub> flux measurements for evaluating a high-resolution urban CO<sub>2</sub> emissions inventory

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Source decomposition of eddy-covariance CO<sub>2</sub> flux measurements  
for evaluating a high-resolution urban CO<sub>2</sub> emissions inventory

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Supplementary material for this article is available [online](#)

## Abstract

We present the comparison of source-partitioned CO<sub>2</sub> flux measurements with a high-resolution urban CO<sub>2</sub> emissions inventory (Hestia). Tower-based measurements of CO and <sup>14</sup>C are used to partition net CO<sub>2</sub> flux measurements into fossil and biogenic components. A flux footprint model is used to quantify spatial variation in flux measurements. We compare the daily cycle and spatial structure of Hestia and eddy-covariance derived fossil fuel CO<sub>2</sub> emissions on a seasonal basis. Hestia inventory emissions exceed the eddy-covariance measured emissions by 0.36 μmol m<sup>-2</sup> s<sup>-1</sup> (3.2%) in the cold season and 0.62 μmol m<sup>-2</sup> s<sup>-1</sup> (9.1%) in the warm season. The daily cycle of fluxes in both products matches closely, with correlations in the hourly mean fluxes of 0.86 (cold season) and 0.93 (warm season). The spatially averaged fluxes also agree in each season and a persistent spatial pattern in the differences during both seasons that may suggest a bias related to residential heating emissions. In addition, in the cold season, the magnitudes of average daytime biological uptake and nighttime respiration at this flux site are approximately 15% and 27% of the mean fossil fuel CO<sub>2</sub> emissions over the same time period, contradicting common assumptions of no significant biological CO<sub>2</sub> exchange in northern cities during winter. This work demonstrates the effectiveness of using trace gas ratios to adapt eddy-covariance flux measurements in urban environments for disaggregating anthropogenic CO<sub>2</sub> emissions and urban ecosystem fluxes at high spatial and temporal resolution.

## 1. Introduction

Cities are becoming the focus for formulating and implementing carbon dioxide (CO<sub>2</sub>) emissions mitigation efforts (Bulkeley 2013, Huttyra *et al* 2014, Lee and Koski 2014). Evaluating the effectiveness of

emissions reduction efforts requires accurate CO<sub>2</sub> emissions estimates (Turnbull *et al* 2018, Lauvaux *et al* 2020). Although cities cover only 3% of the global land area, urban areas are home to 55% of the world's population, a proportion that is expected to increase to 68% by 2050 (Chaouad and Verzeroli 2018).

Overall, more than 70% of global fossil fuel CO<sub>2</sub> (CO<sub>2</sub>ff) emissions are from urban areas (Edenhofer *et al* 2015). Efforts to assess and mitigate CO<sub>2</sub> emissions can provide benefits for urban sustainability and balanced economic growth (Hsu *et al* 2019).

Urban areas are consistently reported as a net source of CO<sub>2</sub> emissions (Velasco and Roth 2010). The eddy-covariance technique has been applied to measure urban CO<sub>2</sub> emissions in different cities for about two decades (Grimmond *et al* 2002, Nemitz *et al* 2002, Vogt *et al* 2006, Christen *et al* 2011, Järvi *et al* 2012, Christen 2014, Lietzke *et al* 2015, Ao *et al* 2016, Helfter *et al* 2016, Park and Schade 2016, Björkegren and Grimmond 2018). The attribution of urban CO<sub>2</sub> flux measurements is challenging due to the spatial heterogeneity, mixed emission sources and sinks, and limited spatial coverage of flux measurements (Aubinet *et al* 2012). Although most previous studies focus on the observed net CO<sub>2</sub> flux, a few studies attempt to partition flux measurements into fossil and biogenic components accounting for the temporal and spatial variability of the multiple sources and sinks. Menzer and McFadden (2017) modeled fossil CO<sub>2</sub> emissions based on winter data and extrapolated them to the growing season to estimate biogenic fluxes. Ishidoya *et al* (2020) demonstrated partitioning of CO<sub>2</sub> fluxes into liquid and gaseous fossil components using O<sub>2</sub> and CO<sub>2</sub> measurements. Sugawara *et al* (2021) used a nearby tower to estimate the biogenic component of a total CO<sub>2</sub> flux measurement.

Quantification of anthropogenic CO<sub>2</sub> emissions is challenging due to the difficulty of separating CO<sub>2</sub>ff emissions from biogenic CO<sub>2</sub> (CO<sub>2</sub>bio) fluxes (Basu *et al* 2020, Miller *et al* 2020). Previous studies demonstrated the feasibility of using <sup>14</sup>C isotope measurements to separate CO<sub>2</sub>ff from CO<sub>2</sub>bio fluxes (Miller *et al* 2012, Turnbull *et al* 2015, Basu *et al* 2016), but flask measurements of <sup>14</sup>C are expensive and discontinuous. Continuous measurements of carbon monoxide (CO) provide another approach to track CO<sub>2</sub>ff emissions (Levin and Karstens 2007, Vogel *et al* 2010, Turnbull *et al* 2011, Silva *et al* 2013, Park and Schade 2016). Uncertainties in the CO to CO<sub>2</sub>ff ratio, which vary as a function of emission sectors, complicate the attribution of urban CO<sub>2</sub> fluxes. The use of <sup>14</sup>C measurements to determine the ratio of CO to CO<sub>2</sub>ff has not yet been applied to eddy covariance flux measurements. We attempt to combine the complementary strengths of CO and <sup>14</sup>C to decompose net CO<sub>2</sub> flux measurements, and use the partitioned CO<sub>2</sub>ff emissions to evaluate a high-resolution emissions inventory.

Emissions inventories use activity data to aggregate urban CO<sub>2</sub>ff emissions (Boden *et al* 2009, Gurney *et al* 2009, Olivier and Janssens-Maenhout 2012), but the differences among inventories are sizeable (Gately and Hutrya 2017, Oda *et al* 2019, Gurney *et al* 2020).

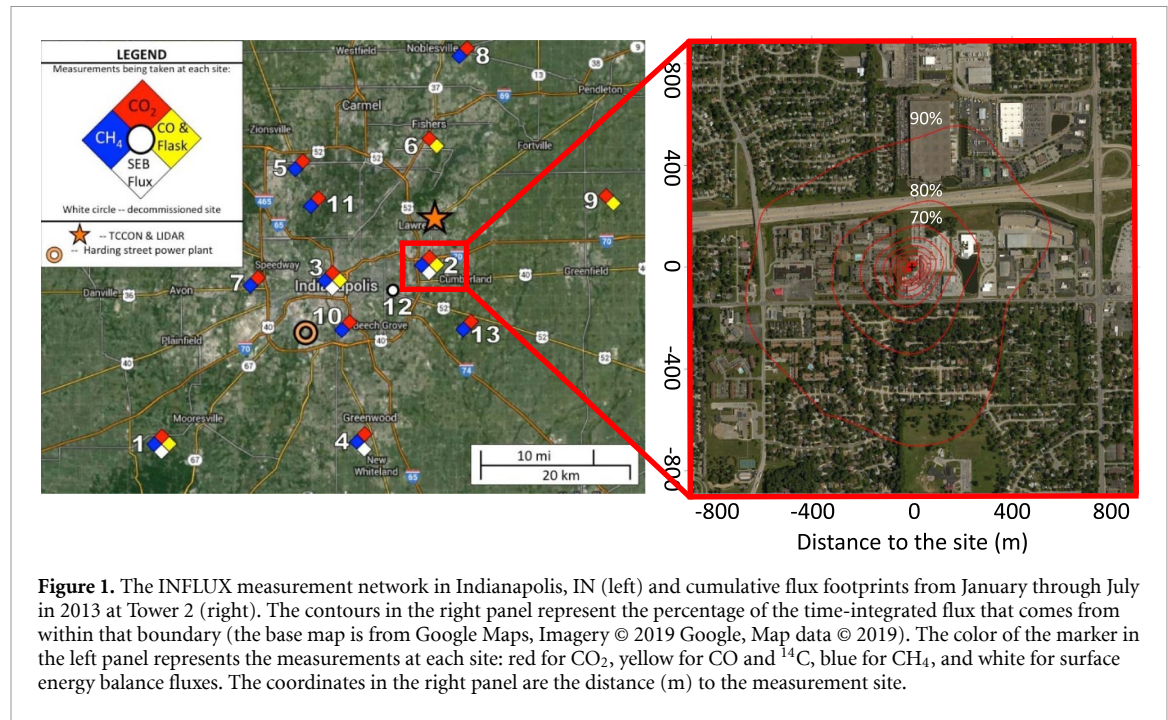
Atmospheric inversions use inventories as prior estimates of emissions and optimize the emissions using atmospheric CO<sub>2</sub> mole fraction observations (Bréon *et al* 2015, Lauvaux *et al* 2016, 2020, Stauder *et al* 2016, Turner *et al* 2016, Kunik *et al* 2019). Two substantial sources of uncertainty in inverse estimates of urban CO<sub>2</sub>ff emissions are uncertain CO<sub>2</sub>bio fluxes and unknown error characteristics in emissions inventories (Wu *et al* 2018). The Hestia emissions inventory (Gurney *et al* 2012) was developed in part to support the Indianapolis Flux Experiment (INFLUX) and uses energy consumption, population density, and traffic data to quantify CO<sub>2</sub>ff emissions for an entire urban landscape at an approximately 200 m and hourly resolution. While excellent agreement between Hestia and atmospheric inversions has been shown over multiple years at the scale of an entire city (Lauvaux *et al* 2020), the high-resolution performance of the Hestia inventory has not yet been evaluated with eddy-covariance flux measurements.

This study compares source-partitioned CO<sub>2</sub> eddy-covariance flux measurements with a high-resolution emissions inventory (Hestia) in a suburban region of Indianapolis, Indiana, USA. We partition the net CO<sub>2</sub> flux measurements into CO<sub>2</sub>ff and CO<sub>2</sub>bio components using a flux-gradient relationship (Stull 2012) and atmospheric CO measurements. <sup>14</sup>C isotope measurements are used to estimate the CO to CO<sub>2</sub>ff ratio and reduce the uncertainty in the flux decomposition. The source decomposition methods are similar to those used by Ishidoya *et al* (2020) and Sugawara *et al* (2021). In addition, we use a flux footprint model (Kljun *et al* 2004, 2015) to match each flux measurement in space and time with the Hestia inventory to provide a direct comparison of independent estimates of anthropogenic CO<sub>2</sub> emissions at high spatial and temporal resolution. This is, to our knowledge, the first such comparison of these innovative and independent assessments of high-resolution urban CO<sub>2</sub> emissions, and is timely given the growing interest in monitoring the impact of urban systems on atmospheric composition.

## 2. Data and methods

### 2.1. Site descriptions and atmospheric CO<sub>2</sub> flux measurements

The INFLUX observation network (Davis *et al* 2017) measures atmospheric CO<sub>2</sub> and CO mole fractions, and net CO<sub>2</sub> fluxes in and around Indianapolis, IN (figure 1). The locations, sampling heights and measurements at these sites are described by Miles *et al* (2017) and the instrument performance is described by Richardson *et al* (2017). <sup>14</sup>C isotope measurements, collected weekly, are used to evaluate CO to CO<sub>2</sub>ff ratios using methods described by Turnbull *et al* (2015).



**Figure 1.** The INFLUX measurement network in Indianapolis, IN (left) and cumulative flux footprints from January through July in 2013 at Tower 2 (right). The contours in the right panel represent the percentage of the time-integrated flux that comes from within that boundary (the base map is from Google Maps, Imagery © 2019 Google, Map data © 2019). The color of the marker in the left panel represents the measurements at each site: red for CO<sub>2</sub>, yellow for CO and <sup>14</sup>C, blue for CH<sub>4</sub>, and white for surface energy balance fluxes. The coordinates in the right panel are the distance (m) to the measurement site.

Since the flux decomposition requires atmospheric measurements of CO<sub>2</sub> and CO mole fractions at different heights as well as <sup>14</sup>C isotope measurements, the need for multiple observational datasets limits the time and location for which we have available data. In total, there are seven months (January through July, 2013) that include all of these data sets (atmospheric measurements of CO<sub>2</sub> and CO mole fractions, <sup>14</sup>C isotope, and CO<sub>2</sub> flux) available at Tower 2 (39.7978° N, 86.0183° W), which is located in a heterogeneous suburban environment (figures 1 and S1). There is a highway to the north, urban vegetation to the south, and neighborhoods with detached houses. The heterogeneous surroundings present a good test of our ability to partition net CO<sub>2</sub> flux measurements into fossil and biogenic components and to use flux footprint analyses to compare the spatial and temporal heterogeneity of source-specific flux data and the Hestia inventory.

The flux instrumentation, which includes a sonic anemometer (Campbell Scientific, CSAT-3) and a high-frequency open-path infrared CO<sub>2</sub> sensor (LI-COR Environmental, LI-7500), is mounted at 30 m above ground level (AGL) on Tower 2. The eddy-covariance technique measures the covariance between fluctuations in vertical wind velocity and CO<sub>2</sub> density to detect the integrated exchange of CO<sub>2</sub> between land and atmosphere (Lee *et al* 2004, Foken and Napo 2008, Aubinet *et al* 2012). We use flux calculation and filtering methods recommended by Vickers and Mahrt (1997). We filter out extreme values outside 3.5  $\sigma$  range of the data (0.2% of data are filtered out) and nighttime fluxes during weak turbulence conditions when the friction velocity is less than 0.2 m s<sup>-1</sup> (3.6% of data are filtered out)

(Gu *et al* 2005). Negative fluxes show contributions of photosynthesis to the flux data (figure S2). Based on the similarity of the diurnal variation of net CO<sub>2</sub> flux measurements (figure S3), we define the cold season as January–March (JFM) and the warm season as April–July (AMJJ).

## 2.2. Partitioning fossil and biogenic CO<sub>2</sub> fluxes

To partition fossil and biogenic components from the net CO<sub>2</sub> flux measurements, we apply a flux-gradient method and atmospheric CO measurements. In addition to flux measurements, we also measure CO<sub>2</sub> and CO mole fractions at 10 and 40 m heights AGL at Tower 2 (Miles *et al* 2017). We use the net flux measurement ( $F_{\text{CO}_2}$ ) and vertical gradient in CO<sub>2</sub> mole fraction ( $\nabla C_{\text{CO}_2}$ ) to solve for the eddy diffusivity ( $K$ ):

$$K = -\frac{F_{\text{CO}_2}}{\nabla C_{\text{CO}_2}}, \quad (1)$$

and use that eddy diffusivity and the CO vertical gradient ( $\nabla C_{\text{CO}}$ ) to solve for the CO flux ( $F_{\text{CO}}$ ):

$$F_{\text{CO}} = -K \nabla C_{\text{CO}}. \quad (2)$$

The fossil fuel CO<sub>2</sub> emission ( $F_{\text{CO}_2\text{ff}}$ ) is estimated by combining the CO flux with the emission ratio ( $R$ ) of CO to CO<sub>2</sub>ff:

$$F_{\text{CO}_2\text{ff}} = \frac{F_{\text{CO}}}{R}, \quad (3)$$

and we attribute the difference between the net flux measurement and the partitioned fossil fuel CO<sub>2</sub> emission to the biogenic CO<sub>2</sub> flux ( $F_{\text{CO}_2\text{bio}}$ ):

$$F_{\text{CO}_2\text{bio}} = F_{\text{CO}_2} - F_{\text{CO}_2\text{ff}}. \quad (4)$$



There are three assumptions in this method: (a) turbulent eddies are small enough that local scalar gradients are proportional to turbulent fluxes; (b) CO and CO<sub>2</sub> are subject to the same vertical mixing processes; (c) within the turbulent flux footprint, CO is mainly produced by fossil fuel combustion simultaneously with CO<sub>2</sub>ff emissions. We filter out counter-gradient fluxes, and limit the eddy diffusivity and CO flux within  $3.5\sigma$  range of their estimates to screen out extreme values caused by tiny denominators. Human respiration, which would appear in this decomposition as a biological flux, is estimated based on the population density of Indianapolis (896 people km<sup>-2</sup> in the year 2013) multiplied by a typical emission rate of 942 gCO<sub>2</sub> person<sup>-1</sup> day<sup>-1</sup> (Prairie and Duarte 2007).

The emission ratio of CO to CO<sub>2</sub>ff is estimated from flask measurements of <sup>14</sup>C and CO measurements (Turnbull *et al* 2015). The urban CO and <sup>14</sup>C enhancements are estimated by the differences between Tower 2 and upwind background sites (Tower 1 or 9 depending on the wind direction). The median and mean values of CO to CO<sub>2</sub>ff ratios estimated from these enhancements are 9.52 and 8.98 ppb ppm<sup>-1</sup> (cold season) and 9.13 and 9.02 ppb ppm<sup>-1</sup> (warm season) (figure S4). We use 9 ppb ppm<sup>-1</sup> as an approximate value to infer CO<sub>2</sub>ff emissions. To test the uncertainty of using different ratios on the flux decomposition, we vary the emission ratio to 11 and 7 ( $9 \pm 2$ ) ppb ppm<sup>-1</sup>. These are plausible bounds (table 2 in Turnbull *et al* 2015) for this flux site, representing approximately the 70th and 30th percentiles of the values. With a linear relation of the flux decomposition to the emission ratio (equation (3)), this maximum and minimum boundary approach represents our limited confidence in the emission ratio and its uncertainty bounds. A more formal error propagation would suggest more confidence than we have in our estimate of the uncertainty in the emission ratio. In addition, since traffic emissions are likely to have a higher ratio and residential emissions have a smaller ratio, we add another scenario with a CO to CO<sub>2</sub>ff ratio of 15 ppb ppm<sup>-1</sup> for northerly winds from the highway and 7 ppb ppm<sup>-1</sup> for the other wind directions based on sectoral emission ratios estimated by Turnbull *et al* (2015).

### 2.3. Flux footprint and emissions inventory

A flux footprint, which is defined as the contributing area upwind from the measurement site (Leclerc and Foken 2014), is essential to account for the spatial heterogeneity of emission sources. We use a two-dimensional flux footprint model (<https://footprint.kljun.net/>) (Kljun *et al* 2004, 2015) to match with the Hestia inventory and estimate the emissions predicted by the inventory at the tower location. Flux footprints were computed with a spatial resolution of approximately 2 m. The size

of footprint depends on measurement height, surface roughness, and atmospheric thermal stability. The footprint will increase with an increase in measurement height, with a decrease in surface roughness, and with an increase in atmospheric thermal stability (Burba and Anderson 2010). Tower-based measurements of wind field and boundary layer characteristics are used to estimate the input parameters of the flux footprint model (measurement height above displacement height, roughness length, Obukhov length, friction velocity, mean wind speed, boundary layer height, standard deviation (SD) of lateral velocity fluctuations). The displacement height and roughness length are estimated as 6 and 0.45 m, respectively. The displacement height is estimated to be 0.7 times the local mean building and tree heights (Weng *et al* 2013) and the roughness length is computed from the mean wind and momentum fluxes measured at 30 m AGL (Drew *et al* 2013, Kent *et al* 2017). We estimate the flux footprint ( $f$ ) for each hourly flux measurement. After interpolating the Hestia inventory to the coordinates of each flux footprint, we weight the hourly Hestia emissions ( $Q_H$ ) with the spatially-resolved fractional flux contributions ( $f$ ) at the same time and sum over the domain of flux footprint ( $R$ ) to produce a spatially-weighted estimate of the Hestia flux that would be measured at the tower ( $F_H$ ):

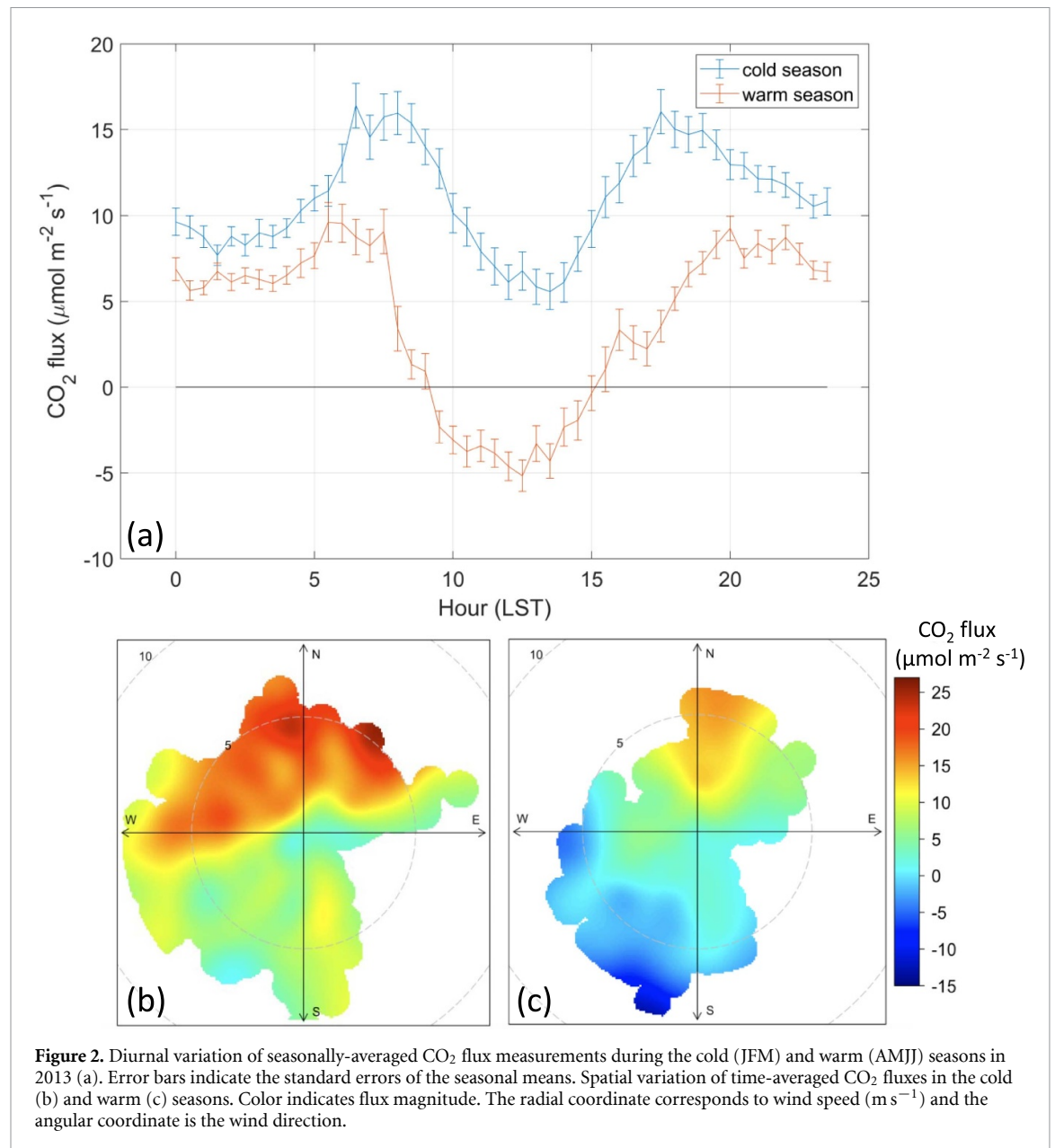
$$F_H = \sum_{i=1}^R Q_H(x_i, y_i) f(x_i, y_i) \delta x \delta y. \quad (5)$$

The emissions predicted by the Hestia inventory at the tower ( $F_H$ ) are compared with the partitioned CO<sub>2</sub>ff flux measurements ( $F_{\text{CO}_2\text{ff}}$  in equation (3)).

## 3. Results

Net CO<sub>2</sub> flux measurements, decomposed as a function of time and space, behave as expected given the environment surrounding the tower. Observed CO<sub>2</sub> emissions are larger in the cold season than the warm season (figure 2(a)), perhaps due to increased emissions from building heating around the tower (figures 1 and S1). In the cold season, there are two prominent peaks in emissions likely corresponding to peaks in traffic volume during rush hours. In the warm season, CO<sub>2</sub>ff emissions are mixed with photosynthesis and respiration from urban vegetation within the flux footprints. The daytime photosynthetic uptake of CO<sub>2</sub> indicates the role of urban vegetation. The data show high emissions from the north, and lower emissions or net uptake from the south (figures 2(b) and (c)), consistent with the highway to the north and urban vegetation to the south of the tower.

Partitioning of the net observed CO<sub>2</sub> fluxes into fossil and biogenic components yields plausible temporal behavior of these flux components (figure 3). While smaller than the estimated CO<sub>2</sub>ff emissions,

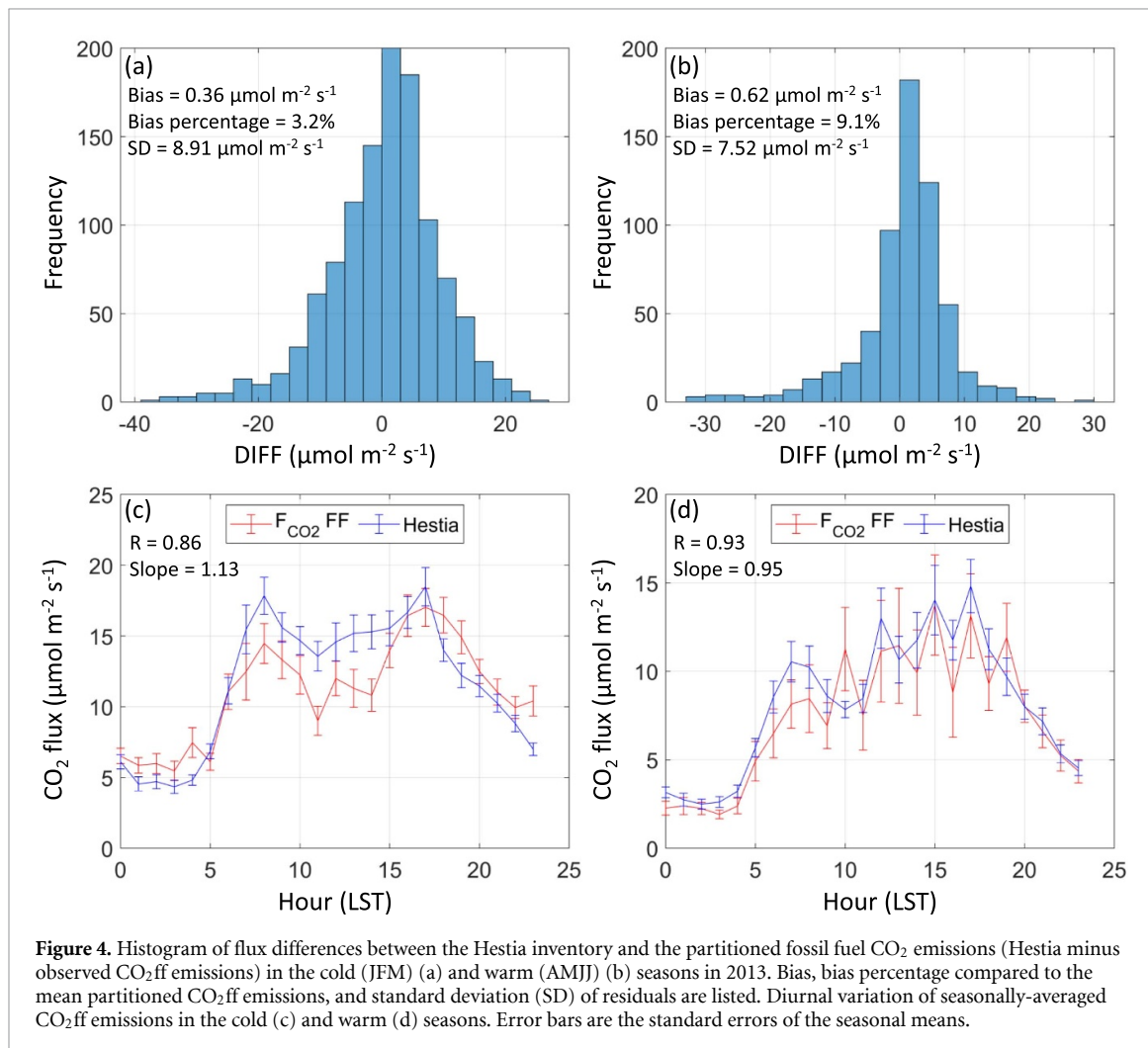
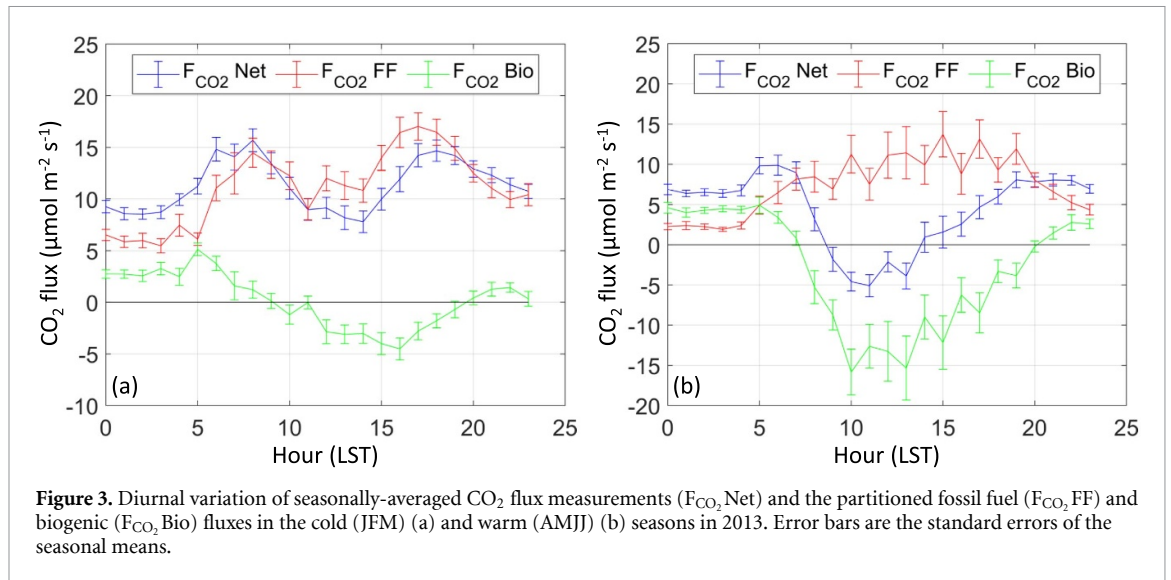


**Figure 2.** Diurnal variation of seasonally-averaged CO<sub>2</sub> flux measurements during the cold (JFM) and warm (AMJJ) seasons in 2013 (a). Error bars indicate the standard errors of the seasonal means. Spatial variation of time-averaged CO<sub>2</sub> fluxes in the cold (b) and warm (c) seasons. Color indicates flux magnitude. The radial coordinate corresponds to wind speed ( $\text{m s}^{-1}$ ) and the angular coordinate is the wind direction.

the magnitude of the cold season daytime (9 to 20 LST) averaged biological uptake is 15% of the mean CO<sub>2</sub>ff emissions over the same time period and the ecosystem respiration averaged over nighttime (21 to 8 LST) is 27% of the mean nighttime CO<sub>2</sub>ff emissions. These are non-negligible flux magnitudes that need to be considered to obtain accurate CO<sub>2</sub>ff emissions (figure 3(a)). Human respiration is estimated to be  $0.22 \mu\text{mol m}^{-2} \text{s}^{-1}$ , which would contribute about 10% of the average nighttime CO<sub>2</sub>bio fluxes in the cold season. A typical pattern of ecosystem fluxes emerges in the warm season (figure 3(b)). The warm season CO<sub>2</sub>bio fluxes are equal in amplitude to the CO<sub>2</sub>ff emissions, emphasizing the importance of accounting for CO<sub>2</sub>bio fluxes in attempts to quantify urban CO<sub>2</sub>ff emissions. The error bars are the standard errors of the seasonal means, which represent a mixture of day-to-day variability, random measurement errors, and uncertainty in the

flux decomposition using a typical emission ratio ( $9 \text{ ppb ppm}^{-1}$ ). We will examine the impacts of using different ratios on the flux decomposition.

The seasonally-averaged partitioned CO<sub>2</sub>ff emissions estimates show remarkable similarity to the Hestia inventory when matched in space and time using the flux footprint model. Seasonal-mean CO<sub>2</sub>ff emissions differ (Hestia minus observed CO<sub>2</sub>ff emissions) by  $0.36 \mu\text{mol m}^{-2} \text{s}^{-1}$  (3.2% of the mean partitioned CO<sub>2</sub>ff emissions) in the cold season (figure 4(a)) and  $0.62 \mu\text{mol m}^{-2} \text{s}^{-1}$  (9.1% of the mean partitioned CO<sub>2</sub>ff emissions) in the warm season (figure 4(b)). The corresponding SDs of the residuals are  $8.91$  and  $7.52 \mu\text{mol m}^{-2} \text{s}^{-1}$ , which include random errors in the flux measurements. The temporal patterns of seasonally-averaged Hestia and the partitioned CO<sub>2</sub>ff emissions also agree remarkably well (figures 4(c) and (d)). The correlation coefficients of the diurnal variations are 0.86

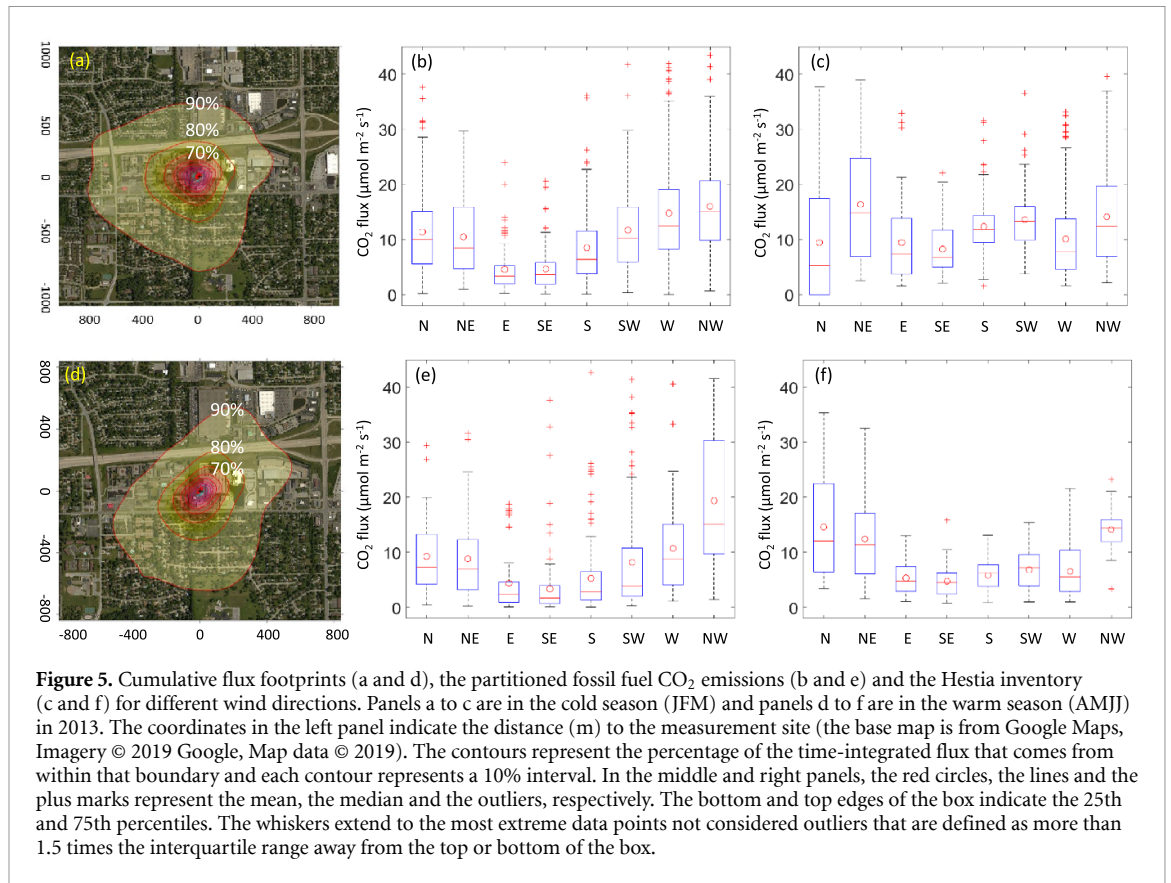


(cold season) and 0.93 (warm season), and the slopes are 1.13 and 0.95, respectively. The Hestia emissions are smaller during the night and higher during the day compared to the partitioned observations in the cold season (figures 4(c) and S5(a)), and consistently

slightly higher than the partitioned observations in the warm season (figures 4(d) and S5(b)).

We also find similarity in the comparison of eddy-covariance and Hestia  $\text{CO}_2$ ff emissions as a function of wind direction (figure 5). In the cold season, the





**Figure 5.** Cumulative flux footprints (a and d), the partitioned fossil fuel CO<sub>2</sub> emissions (b and e) and the Hestia inventory (c and f) for different wind directions. Panels a to c are in the cold season (JFM) and panels d to f are in the warm season (AMJJ) in 2013. The coordinates in the left panel indicate the distance (m) to the measurement site (the base map is from Google Maps, Imagery © 2019 Google, Map data © 2019). The contours represent the percentage of the time-integrated flux that comes from within that boundary and each contour represents a 10% interval. In the middle and right panels, the red circles, the lines and the plus marks represent the mean, the median and the outliers, respectively. The bottom and top edges of the box indicate the 25th and 75th percentiles. The whiskers extend to the most extreme data points not considered outliers that are defined as more than 1.5 times the interquartile range away from the top or bottom of the box.

**Table 1.** Statistics of flux differences ( $\mu\text{mol m}^{-2} \text{s}^{-1}$ ) between the Hestia inventory and the partitioned fossil fuel CO<sub>2</sub> emissions (Hestia minus observed CO<sub>2</sub>ff emissions) for different wind directions.

	DIFF	N	NE	E	SE	S	SW	W	NW
Cold season (JFM)	Median	−2.00	3.32	2.88	3.45	4.14	3.15	−4.47	−2.14
	Mean	−1.93	5.88	4.88	3.58	3.84	1.89	−4.72	−1.87
	RMSE <sup>a</sup>	10.98	9.27	8.22	5.63	7.45	8.00	10.40	9.06
Warm season (AMJJ)	Median	2.49	3.34	1.92	1.98	0.98	0.42	−2.71	−4.27
	Mean	5.31	3.61	0.92	1.37	0.52	−1.32	−4.17	−5.21
	RMSE	8.24	9.32	5.19	5.54	5.97	8.62	8.47	13.66

<sup>a</sup> Root mean square error.

Hestia emissions are higher than the observed CO<sub>2</sub>ff emissions for all wind directions except the north, west and northwest wind (table 1). A similar pattern exists in the warm season. Since residential buildings lie upwind in the west and northwest wind directions (figures 1 and S1), we infer residential emissions could be the source of this discrepancy.

These results are somewhat sensitive to the choice of CO to CO<sub>2</sub>ff ratio in the flux decomposition. Seasonal-mean flux bias and bias percentage change significantly when the emission ratio varies from 9 ppb ppm<sup>−1</sup> to 11 or 7 ppb ppm<sup>−1</sup> (figure S6 and table S1). Figure S6 shows the impact of plausible ratios on the diurnal cycle of the partitioned CO<sub>2</sub>ff and CO<sub>2</sub>bio fluxes. The lower bound of 7 ppb ppm<sup>−1</sup> increases the CO<sub>2</sub>ff emissions estimate (figure S6(b) and equation (3)),

thus driving the CO<sub>2</sub>bio fluxes down about  $3 \mu\text{mol m}^{-2} \text{s}^{-1}$  in the cold season (figure S6(c) and equation (4)). This would strengthen the finding of daytime photosynthesis. The upper bound ratio of 11 ppb ppm<sup>−1</sup> would increase CO<sub>2</sub>bio fluxes by about  $2 \mu\text{mol m}^{-2} \text{s}^{-1}$  in the cold season, leaving midday fluxes slightly negative and nighttime respiration at about  $4 \mu\text{mol m}^{-2} \text{s}^{-1}$ . Similar results are shown in the warm season (figures S6(e) and S6(f)). The magnitude of the partitioned fluxes varies linearly with the change of emission ratio, but the diurnal cycle is not sensitive to this choice. The scenario with the space-varying emission ratio (15 and 7 ppb ppm<sup>−1</sup>), which may be more realistic than a constant ratio, does not significantly change either the diurnal variation (figure S6) or the bias estimation (table S1) when compared to the default scenario (9 ppb ppm<sup>−1</sup>).

## 4. Conclusions and discussion

The remarkable agreement between the Hestia inventory and the partitioned flux measurements suggests that both methods are able to describe the temporal and spatial variability in urban CO<sub>2</sub>ff emissions at neighborhood scale. Neither approach has yet been cross-validated at such a high spatial and temporal resolution. The flux measurement partitioning is sensitive to the CO to CO<sub>2</sub>ff emission ratio, but the consistency of Hestia and flux data suggests that flask measurements have accurately quantified that ratio. The success of this test suggests that these eddy-covariance flux decomposition methods can be used to quantify source-specific, neighborhood-scale CO<sub>2</sub>ff emissions. Further the successful comparison to Hestia suggests that the algorithms and input data used in the inventory system are accurate and precise even at the fine resolution of the eddy-covariance flux measurements.

This study also shows the promise of using this approach for studying urban ecosystem CO<sub>2</sub> fluxes. Previous work has suggested that the edges found in urban ecosystems lead to fundamentally different behavior of these ecosystems (Reinmann *et al* 2020). These findings are largely based on chamber-scale flux measurements. It is not clear whether or not, when upscaled to spatial domains that integrate across many edges such as a suburban forest, existing ecosystem models and model parameters will suffice in describing urban CO<sub>2</sub>bio fluxes. Current ecosystem models used in urban studies are largely devoid of urban ecosystem flux measurements in either calibration or evaluation due to lack of data (Hardiman *et al* 2017, Wu *et al* 2021). We suggest that the decomposition methods can serve as a new approach for obtaining ecosystem flux data necessary to develop the next generation of urban ecosystem models.

Finally, this study emphasizes the importance of urban ecosystem fluxes, both in the warm (growing) season and the cold (dormant) season. Our results appear to contradict the findings of Turnbull *et al* (2015) who found no net impact of biological CO<sub>2</sub> fluxes on CO<sub>2</sub> enhancements in Indianapolis outside of the growing season. We found the percentage of daytime biological uptake in the cold season is 15% compared to the mean CO<sub>2</sub>ff emissions. Our results are consistent with the flask measurements (figure 5 in Turnbull *et al* 2015) which showed that, for Tower 2, the total CO<sub>2</sub> enhancement in the winter months was 0.8 to 0.9 times the CO<sub>2</sub>ff enhancement, suggesting modest net biological uptake of CO<sub>2</sub> during these months within the city. The flask <sup>14</sup>C-based CO<sub>2</sub>bio enhancement at Tower 2 averaged over the cold season for the three months of this study is −0.37 ppm (table S2) that is about 10% of the estimated fossil CO<sub>2</sub> enhancement (3.6 ppm), consistent with our eddy-covariance

flux measurements. Turnbull *et al* (2015) found no net biological CO<sub>2</sub> contribution to the wintertime enhancements when averaging together four towers including Tower 2. The other towers likely have less influence from urban vegetation based on their position around the city. The importance of growing season biological fluxes has been shown in multiple observational (Turnbull *et al* 2015, Miller *et al* 2020) and inversion (Sargent *et al* 2018, Wu *et al* 2018, Lauvaux *et al* 2020) studies. Uncertainty in biological fluxes has a large impact on inverse flux estimates (Wu *et al* 2018, Lauvaux *et al* 2020). This flux decomposition approach enables evaluation of the modeled ecosystem flux priors using direct urban ecosystem CO<sub>2</sub> flux measurements. Further, a number of studies (Lauvaux *et al* 2016, Heimburger *et al* 2017) have made the reasonable assumption of neglecting CO<sub>2</sub>bio fluxes in the dormant season. This work shows that urban ecosystems in Indianapolis are moderately active even in the cold season. Additional eddy-covariance flux measurements are needed to study the spatial and temporal variations in urban ecosystem CO<sub>2</sub> fluxes.

## Data availability statement

The Hestia emission inventory is available at <http://dx.doi.org/10.18434/T4/1503341>. The eddy-covariance flux measurements and flask data are available at <https://sites.psu.edu/influx/data>. The CO<sub>2</sub> and CO mole fraction measurements are available at <http://dx.doi.org/10.18113/D37G6P>.

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## Conflict of interest

The authors declare no competing interests.

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