Supplementary Information

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3	Assessment of uncertainties of an aircraft-based mass-balance approach for quantifying
4	urban greenhouse gas emissions
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	Flight Date (2011)	Observed CBL depth (m)	Comment
	01 March	525	
	29 April	1110	
	01 June	1310	1 st vertical profile
		1880	2 nd vertical profile
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24 Table S1. Observed depths of the convective boundary layer for the three flight dates.

40	Table S2.	CO ₂ and CH	4 background	averaged over	all altitudes	$(\text{mean} \pm 1 s_m)$), where $1s_m$ is	the
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41 standard deviation of the mean at 95% CL calculated using atmospheric boundary layer statistics

_	Flight Date (2011)	CH ₄ (ppb)	CO ₂ (ppm)
_	01 March	1955.2 ± 1.3	407.4 ± 0.3
	29 April	1896.8 ± 3.2	399.6 ± 0.2
	01 June	1880.8 ± 2.6	392.6 ± 0.5
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42 (Lenschow and Stankov, 1986).

58	Table S3. Hestia (Gurney et al., 2012) and aircraft-based mass balance (Mays et al., 2009)
59	citywide CO_2 emissions flux for 2008 and 2009. Uncertainty in the Hestia county level
60	(equivalent to Indianapolis) fossil fuel CO ₂ emissions is $[-15\%, +20\%]$ at 95% C.I. while
61	uncertainty in the mass balance approach is $\pm 50\%$ (discussion of the precision of the mass
62	balance approach is presented in Section 3.4 in the main text).

Flight Date	Mass Balance CO ₂ Flux (mol s ⁻¹)	Hestia Flux of fossil fuel $CO_2 \pmod{s^{-1}}$
28 March 2008	8080	11222
02 April 2008	2500	9354
14 April 2008	9800	8324
15 April 2008	14000	9308
21 April 2008	6200	6084
28 November 2008	33000	7607
20 December 2008	30000	11552
07 January 2009	8700	12742

Flight	Total No. of	No. of Usebla	Transect	CO. Flux	CU. Flux
Date in		No. of Usable	Height	CO_2 Flux	
2011	Transects	Transects	(m a.g.l.)	$(mol s^{-})$	$(mol s^{-1})$
01 March	4	2	290	17000	170
			360	8700	85
				(12800, 65%) [†]	(130, 65%) [†]
29 April	7	5	200	6200	42
			280	11000	140
			380	16000	190
			480	4600	63
			590	6960	88
				8950 ± 4600	105 ± 60
				(51%)	(58%)
01 June	7	5	210	17900	95
			400	29970	120
			710	30200	190
			1020	23000	260
			1210	21000	270
				24000 ± 5500	190 ± 79
				(23%)	(42%)

Table S4. CO₂ and CH₄ fluxes derived from the single transect method. Also shown are the

70 mean and standard deviation as w	ell as the relative	uncertainty in parenthesis.
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Figure S1. Inflight calibration curves for CO_2 and CH_4 for several flight experiments in 2011.

74 Dashed lines represent the linear least squares fits that were used to adjust the indicated





Figure S2. Comparison of CO₂ (A) and CH₄ (B) concentrations derived from continuous

77 measurements with the cavity ring-down spectrometer (CRDS) and grab sampling with the

78 Programmable Flask Package (PFP).



Figure S3. Frequency distribution of the relative uncertainty between the continuous CRDS
measurements and discrete flask sampling (Figure S2) for (A) CO₂, and (B) CH₄. The relative
uncertainty is defined as percentage difference between the CRDS and flask measurement where
the flask measurement is taken to be the true value.



Figure S4. Vertically-varying background CO₂ and CH₄ for 01 March (A & B), 29 April (C &



- 88 CL.



92 Figure S5. Descending vertical profiles of Potential Temperature and H₂O on (A) 01 March, (B)

93 29 April; and 01 June 2011 (C) before and (D) after the horizontal transects. Broken lines

94 indicate the estimated CBL depths for the three flight experiments.



Figure S6. Flight paths for: Newton County landfill experiments on (A) 16 June 2011 and (B) 03
May 2012 where FODF stands for Fair Oaks Dairy Farm, (C) Twin Bridges LF on 30 August
2012, and (D) Harding Street power plant experiment on 01 June 2012.



98 Figure S6 continued.





Figure S7. Total CO₂ versus fossil fuel CO₂ from flask measurements on 29 April and 01 June,
2011 flight experiments. Filled circles correspond to flask measurements sampled downwind of
the Harding Street Power Plant (HSPP) plume. Radiocarbon CO₂ was not measured on 01
March 2011.



Figure S8. (A) CO_2 and (B) CH_4 observations along the horizontal transects for the various altitudes on 01 March 2011. CBL depth was 525 m. Broken lines represent the minimum and maximum horizontal limits of the city.