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Supplement of

Assessment of uncertainties of an aircraft-based mass balance approach for quantifying urban greenhouse gas emissions

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Table S1. Calculated uncertainties in the analyses parameters used in the sensitivity analyses presented in sections 2.3 (Methods) and 3.3 (Results and Discussion) in the main manuscript. Results of the sensitivity analyses were presented in Table 2 in the main text.

Change (Δ) in Analyses Parameter	01 March 2011	29 April 2011	01 June 2011
Background CO_2 (ppm)[a]	0.3	0.2	0.5
Background CH_4 (ppb)[a]	1.3	3.2	2.6
CBL depth (m)	75	140	204
$\overline{U_{\perp}}$ (ms^{-1})	0.7	0.7	0.7
Background CO_2 due to UHI (ppm)[b]	1	0.2	0.1
Background CH_4 due to UHI (ppb)[b]	7.7	3.5	1.9

[a] The uncertainties in background CO_2 and CH_4 are not constant with altitude. The ΔCO_2 and ΔCH_4 presented in this table are the average uncertainties in the CO_2 and CH_4 backgrounds. See section 2.3 in the main text for detailed discussion as well as Figure S5.

[b] This is the change in the CO_2 and CH_4 backgrounds assuming a 10% change in the urban CBL depth due to the urban heat island effect. See Table S5 for details.

Table S2. Observed depths of the convective boundary layer for the three flight dates.

Flight Date (2011)	Observed CBL depth (m)	Comment
01 March	525	
29 April	1110	
01 June	1310	1st vertical profile
	1880	2nd vertical profile

Table S3. CO_2 and CH_4 background averaged over all altitudes (mean ± $1s_m$), where $1s_m$ is the standard deviation of the mean at 95% CL calculated using atmospheric boundary layer statistics (Lenschow and Stankov, 1986).

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	1880.6 ± 2.6	392.6 ± 0.5

Table S4. Hestia (Gurney et al., 2012) and aircraft-based mass balance (Mays et al., 2009) citywide CO_2 emissions flux for 2008 and 2009. Uncertainty in the Hestia county level (equivalent to Indianapolis) fossil fuel CO_2 emissions is [-15%, +20%] at 95% C.I. while uncertainty in the mass balance approach is ±50% (discussion of the precision of the mass 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 (mol 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

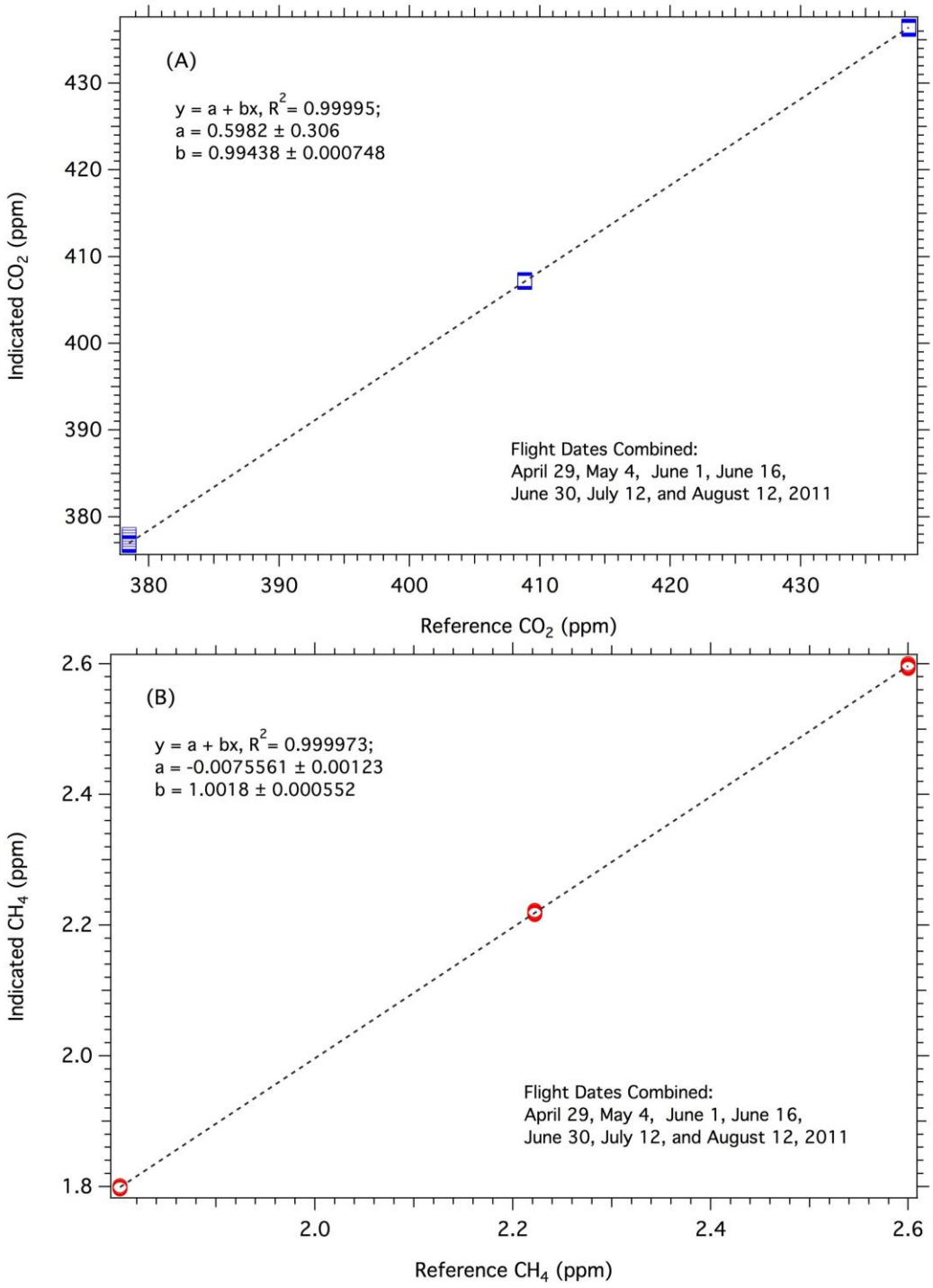
Table S5. Sensitivity analysis of the CO_2 and CH_4 emission rate as a function of change in CO_2 and CH_4 background molar ratios due to enhanced heat island boundary layer depths. We assumed a 10% increase in BL depth consistent with our measurements and that of Angevine et al. (2003).

Flight Date in 2011	BL depth (m)	Bkgnd CO_2 (ppm)	Bkgnd CH_4 (ppb)	Ave CO_2 in Free trop (ppm)	Ave CH_4 in Free trop (ppb)	Adjusted CO_2 bkgnd (ppm)	Adjusted CH_4 bkgnd (ppb)	ΔCO_2 (ppm)	ΔCH_4 (ppb)	% Change in CO_2 Flux	% Change in CH_4 Flux
01 Mar	525	407.4 ± 0.3	1955.2 ± 1.3	397.7 ± 1.0	1876.6 ± 7.3	406.4	1947.3	1	7.7	57	57
29 Apr	1110	399.6 ± 0.2	1896.8 ± 3.2	397.6 ± 0.5	1861.6 ± 6.3	399.4	1893.3	0.2	3.5	22	35
01 Jun	1720	392.6 ± 0.5	1880.8 ± 2.6	393.3 ± 0.6	1862.2 ± 6.6	392.7	1878.9	0.1	1.9	6.3	23
							Average:	**0.4**	**4.4**	**28**	**38**

Table S6. CO_2 and CH_4 fluxes derived from the single transect method. Also shown are the mean and standard deviation as well as the relative uncertainty in parenthesis.

Flight Date in 2011	Total No. of Transects	No. of Usable Transects	Transect Height (m a.g.l.)	CO_2 Flux (mol s^{-1})	CH_4 Flux (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%)

† Mean and percentage difference of the fluxes from the two transects.



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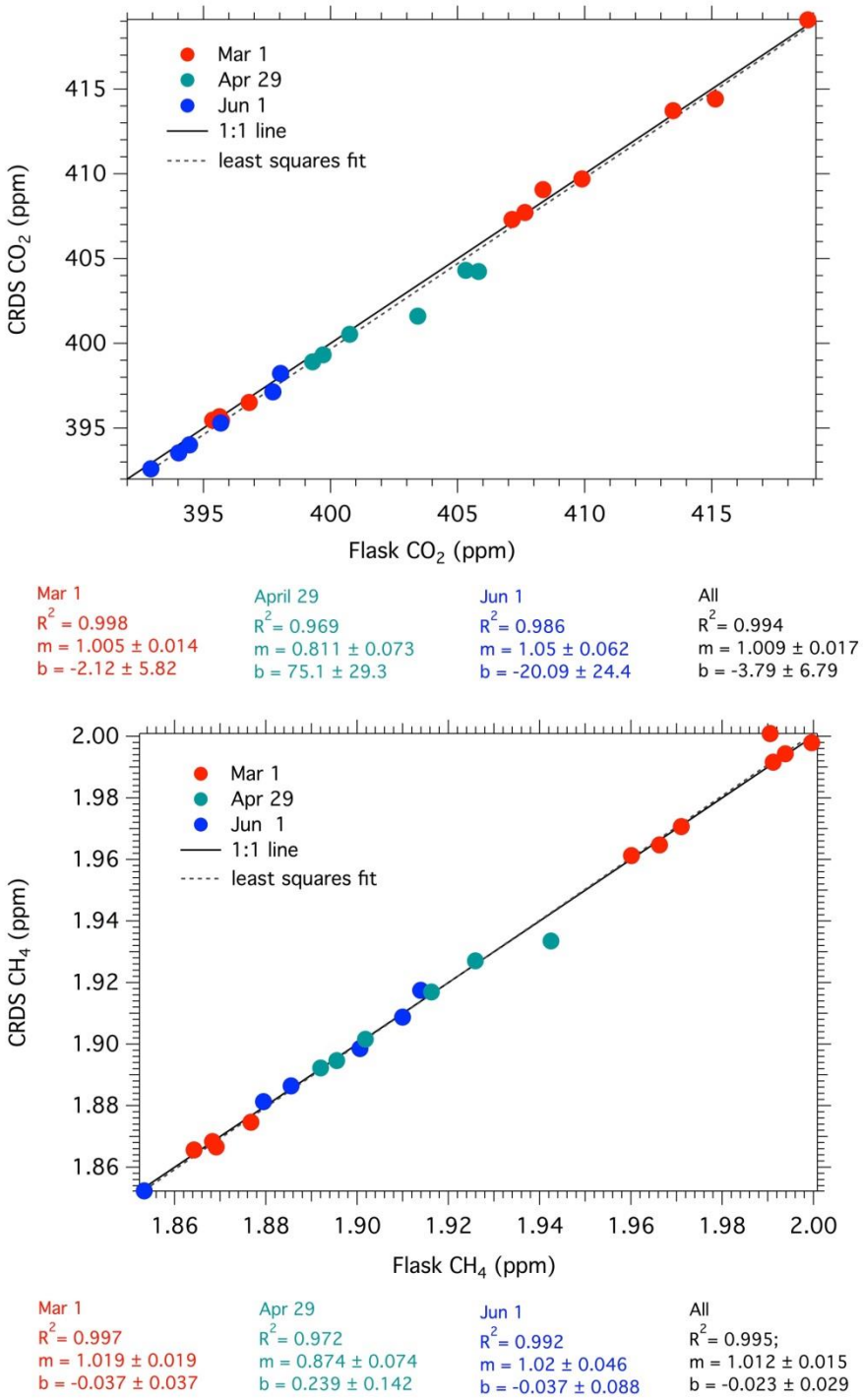
94 Figure S1. Inflight calibration curves for CO_2 and CH_4 for several flight experiments in 2011.

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

96 concentrations to the true CO_2 and CH_4 concentrations.



97 Figure S2. Comparison of CO_2 (A) and CH_4 (B) concentrations derived from continuous
98 measurements with the cavity ring-down spectrometer (CRDS) and grab sampling with the
99 Programmable Flask Package (PFP).

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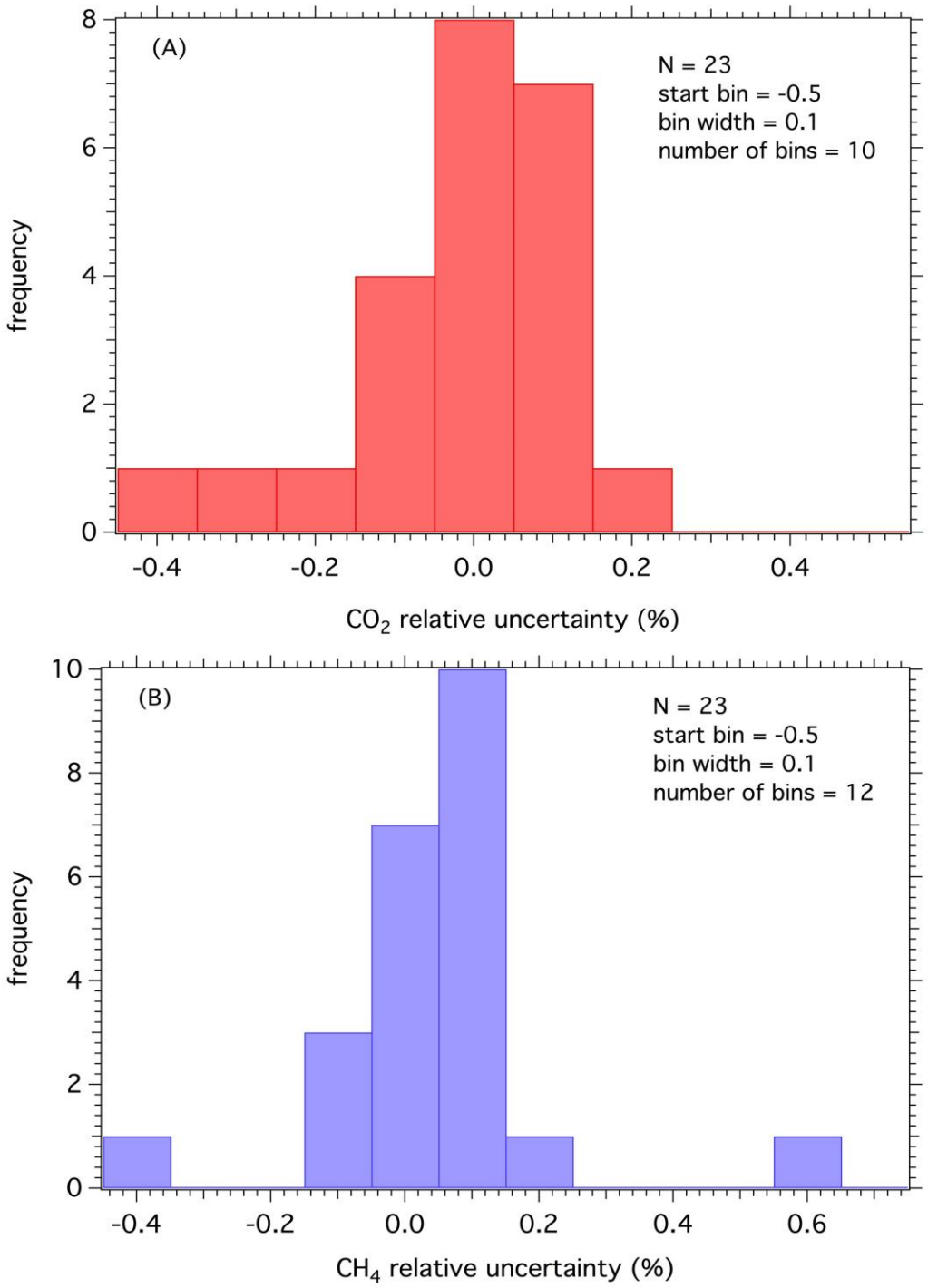
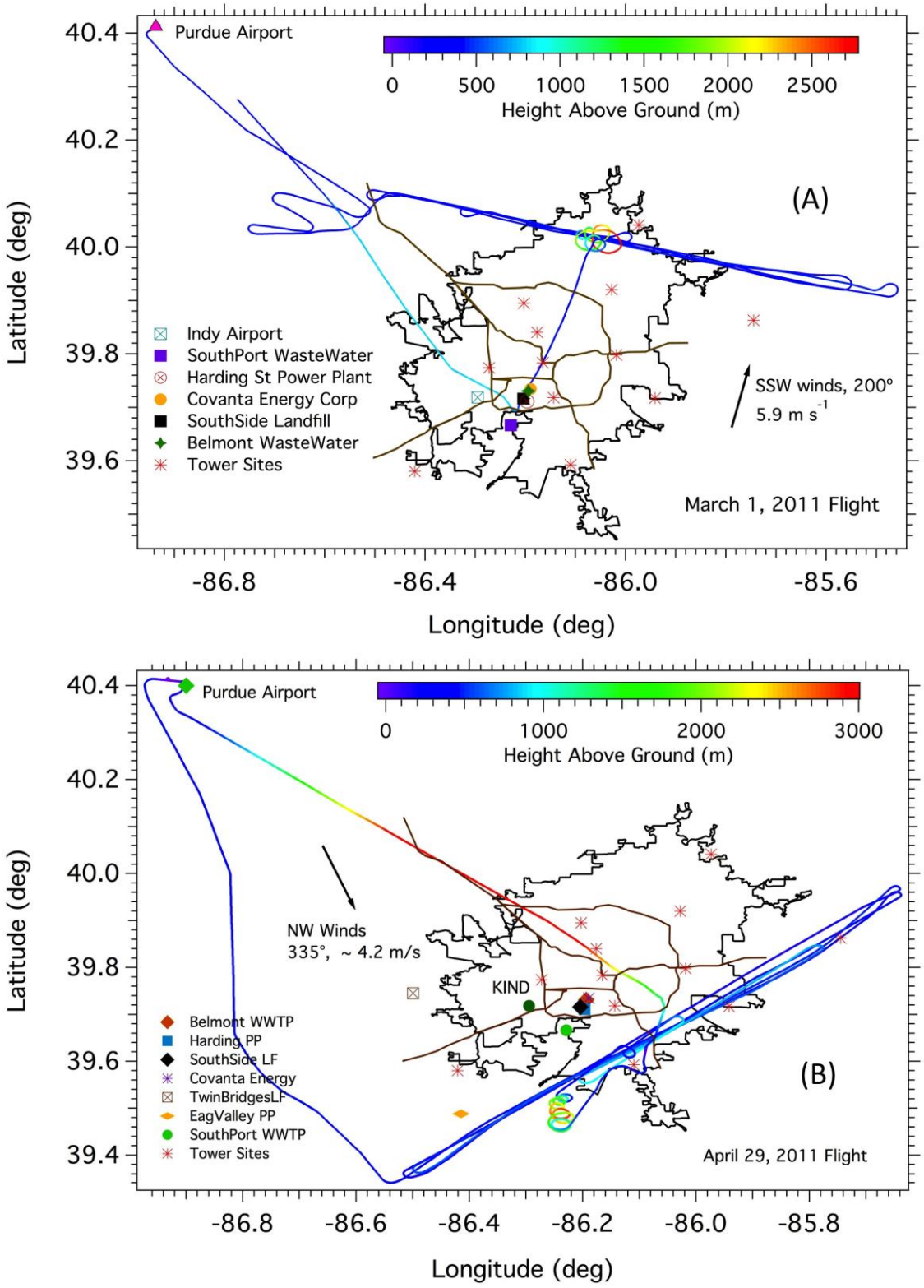


Figure S3. Frequency distribution of the relative uncertainty between the continuous CRDS measurements and discrete flask sampling (Figure S2) for (A) CO_2, and (B) CH_4. The relative uncertainty is defined as percentage difference between the CRDS and flask measurement where the flask measurement is taken to be the true value.



105 Figure S4. Experimental flight paths on (A) 01 March and (B) 29 April 2011 Indianapolis flight

106 experiments.

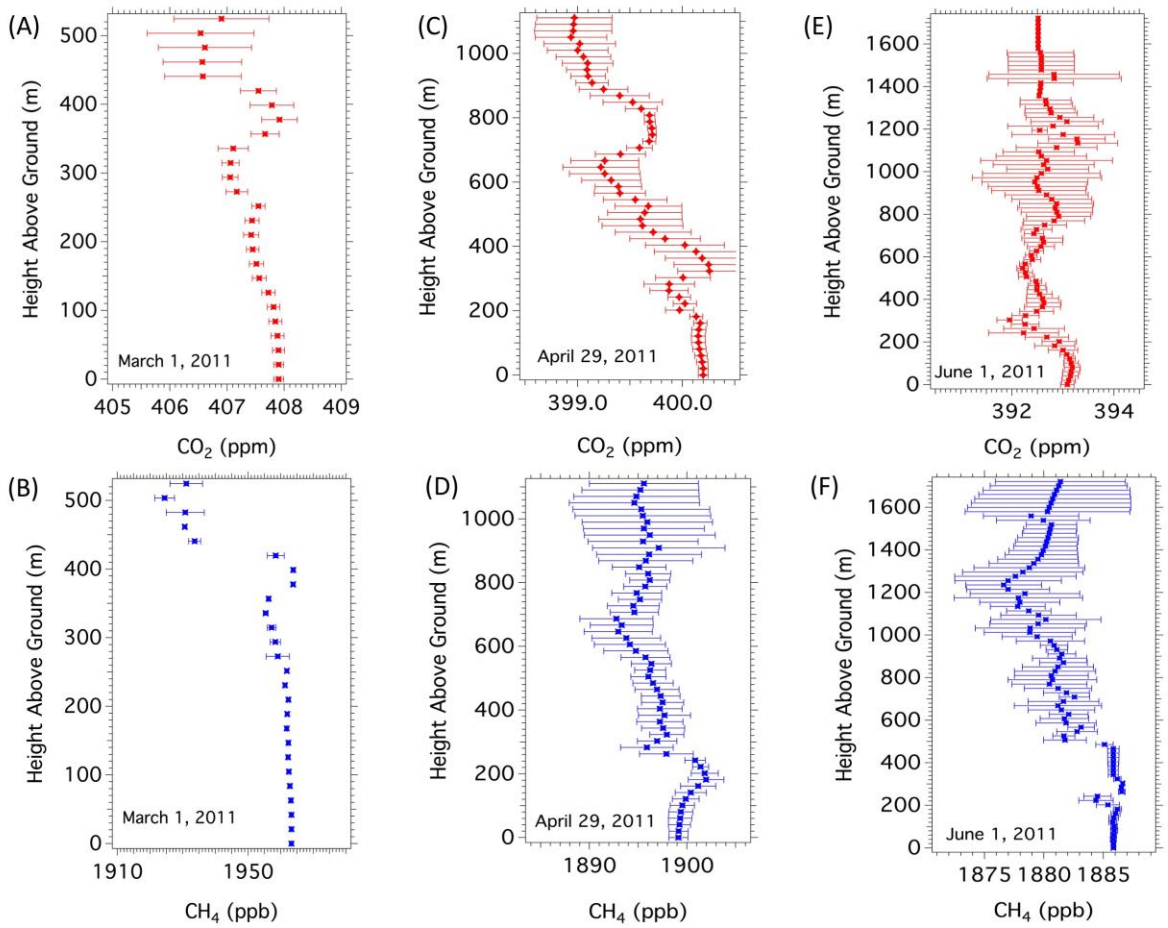
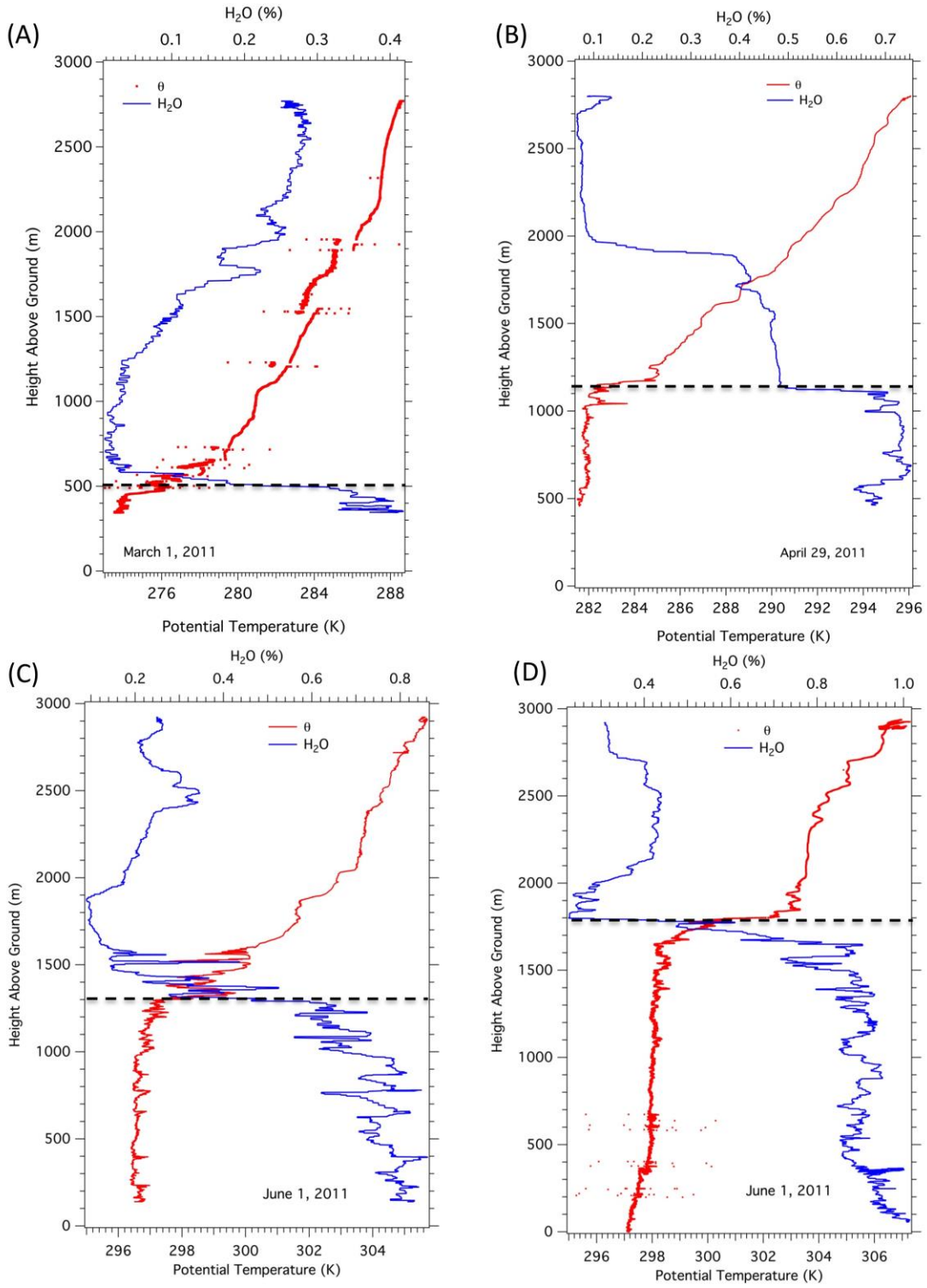


Figure S5. Vertically-varying background CO_2 and CH_4 for 01 March (A & B), 29 April (C & D), and 01 June (E & F) 2011. Error bars represent the standard deviation of the mean at 95% CL.



116 Figure S6. Descending vertical profiles of Potential Temperature and H_2O on (A) 01 March, (B)

117 29 April; and 01 June 2011 (C) before and (D) after the horizontal transects. Broken lines

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

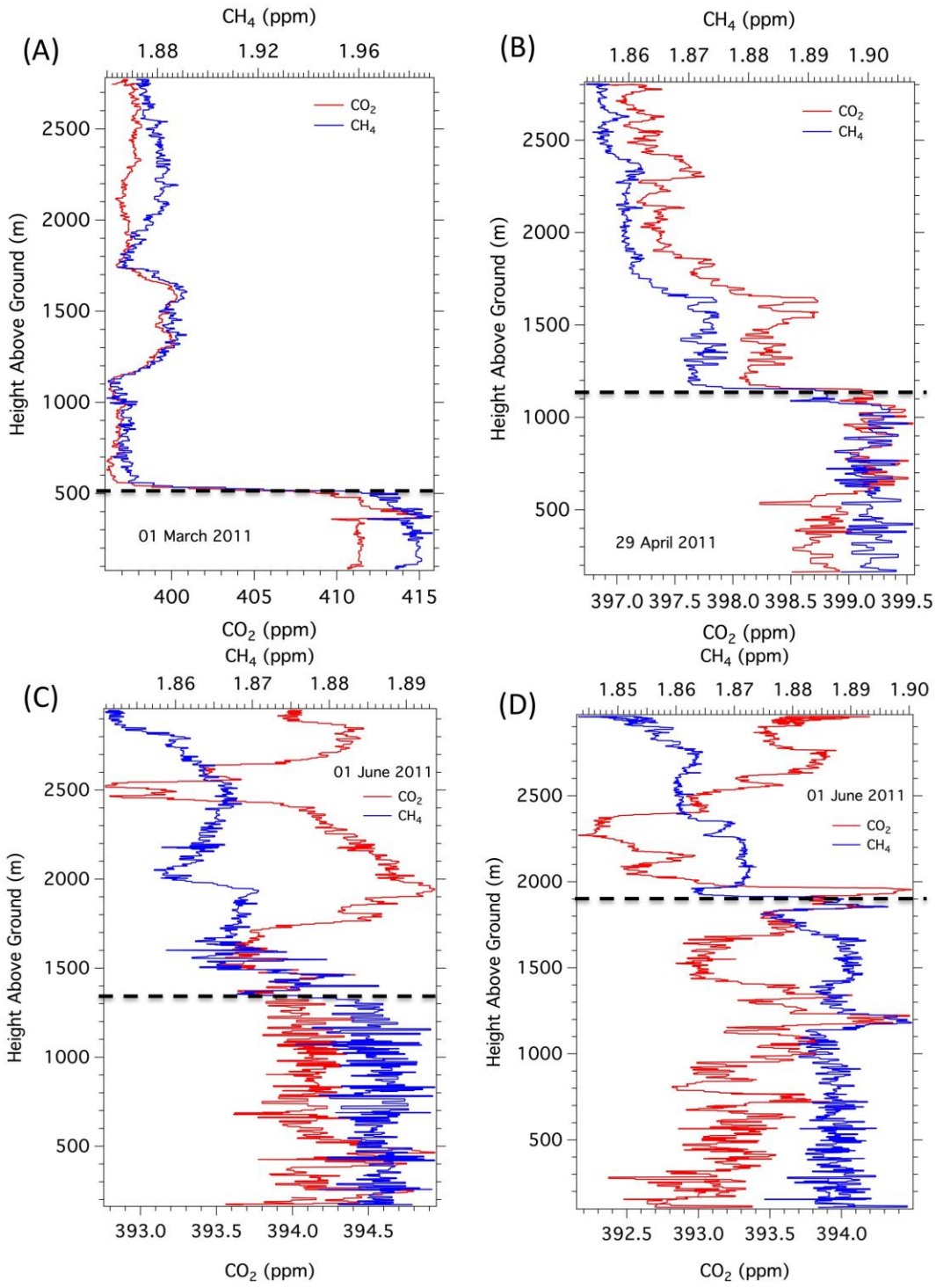
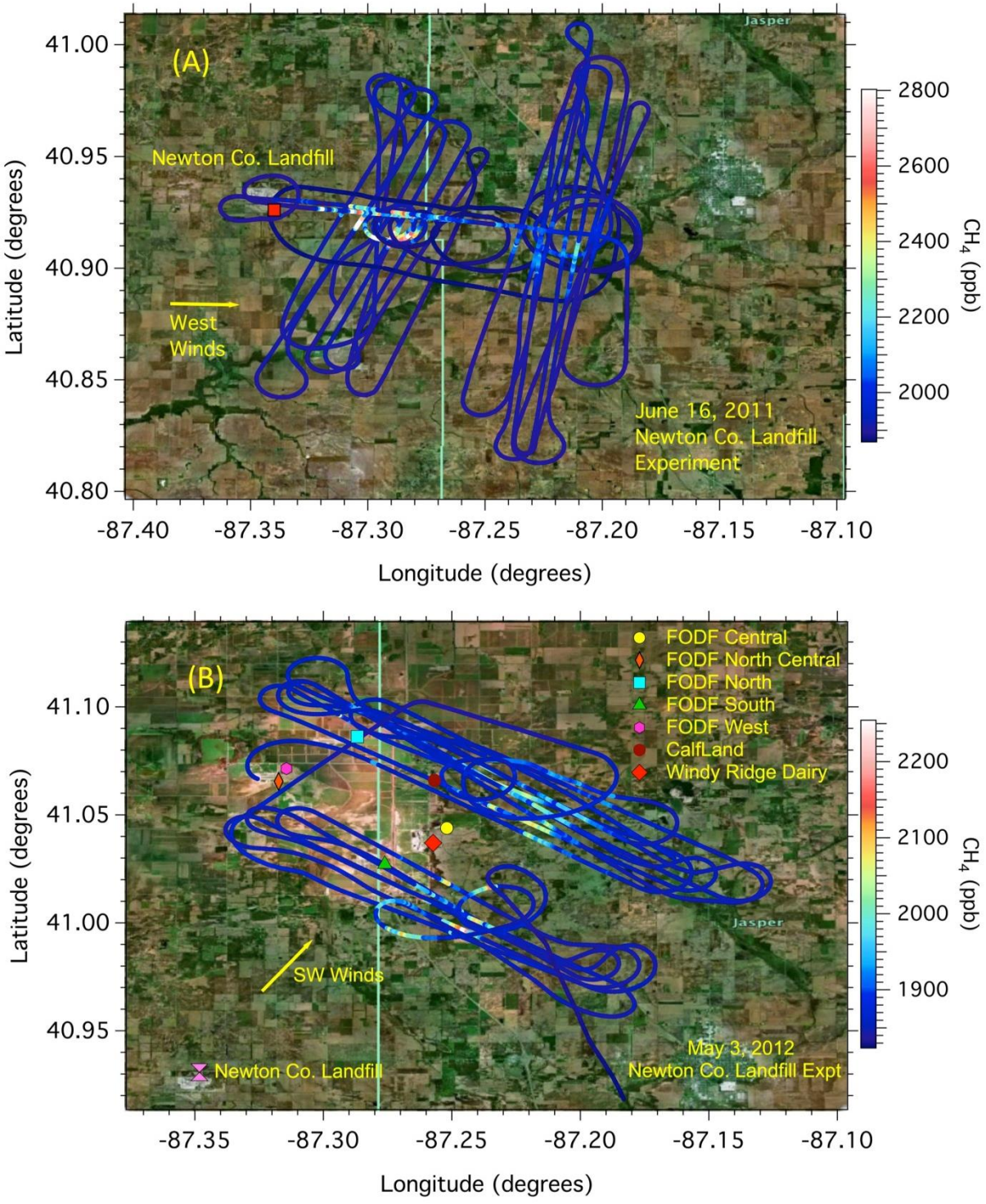


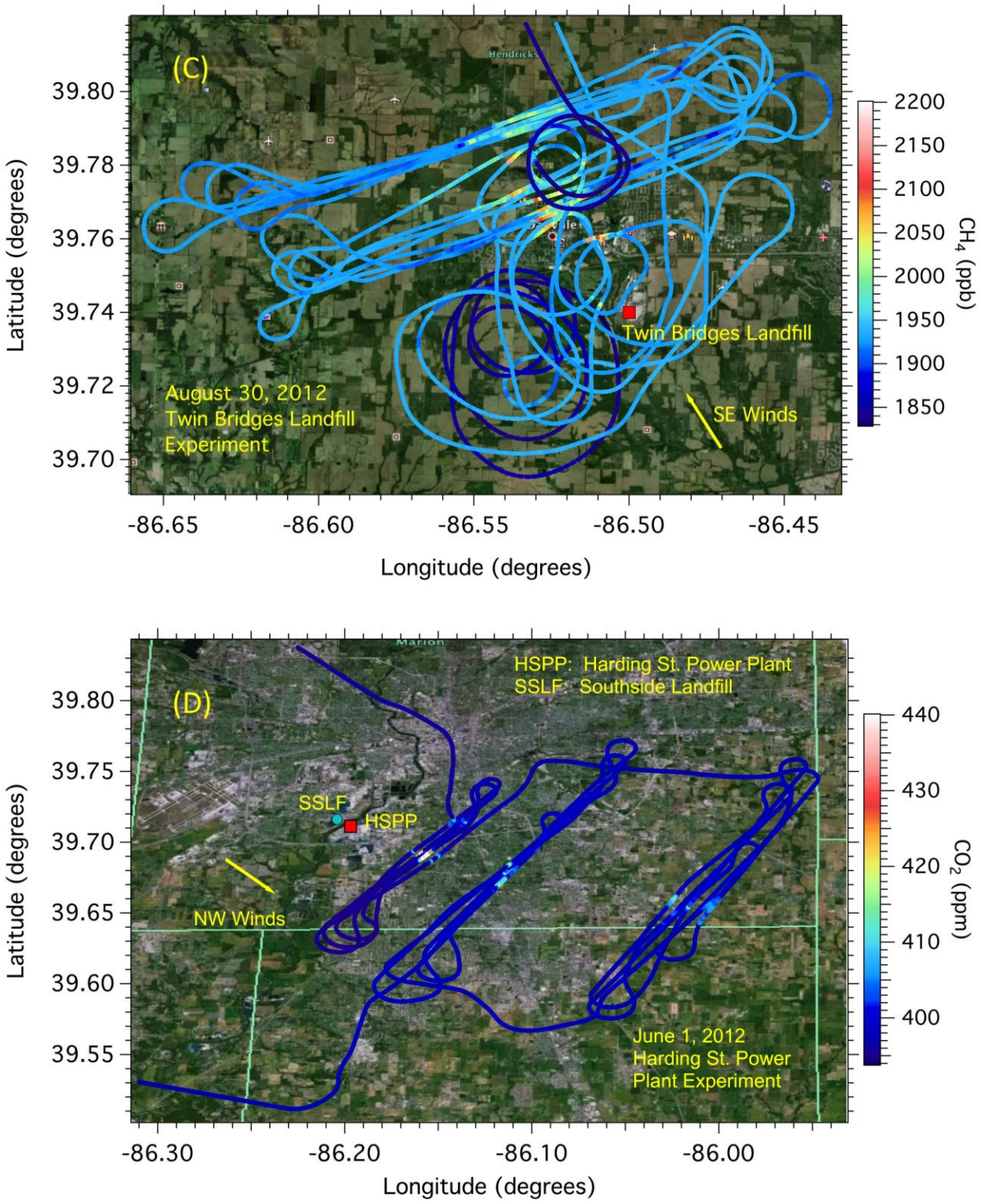
Figure S7. Ascending vertical profiles of CO_2 and CH_4 (A) 01 March, (B) 29 April; and 01 June 2011 (C) before and (D) after the horizontal transects. Broken lines indicate the estimated CBL depths for the three flight experiments.



123 Figure S8. Flight paths for: Newton County landfill experiments on (A) 16 June 2011 and (B) 03

124 May 2012 where FODF stands for Fair Oaks Dairy Farm, (C) Twin Bridges LF on 30 August

125 2012, and (D) Harding Street power plant experiment on 01 June 2012.



126 Figure S8 continued.

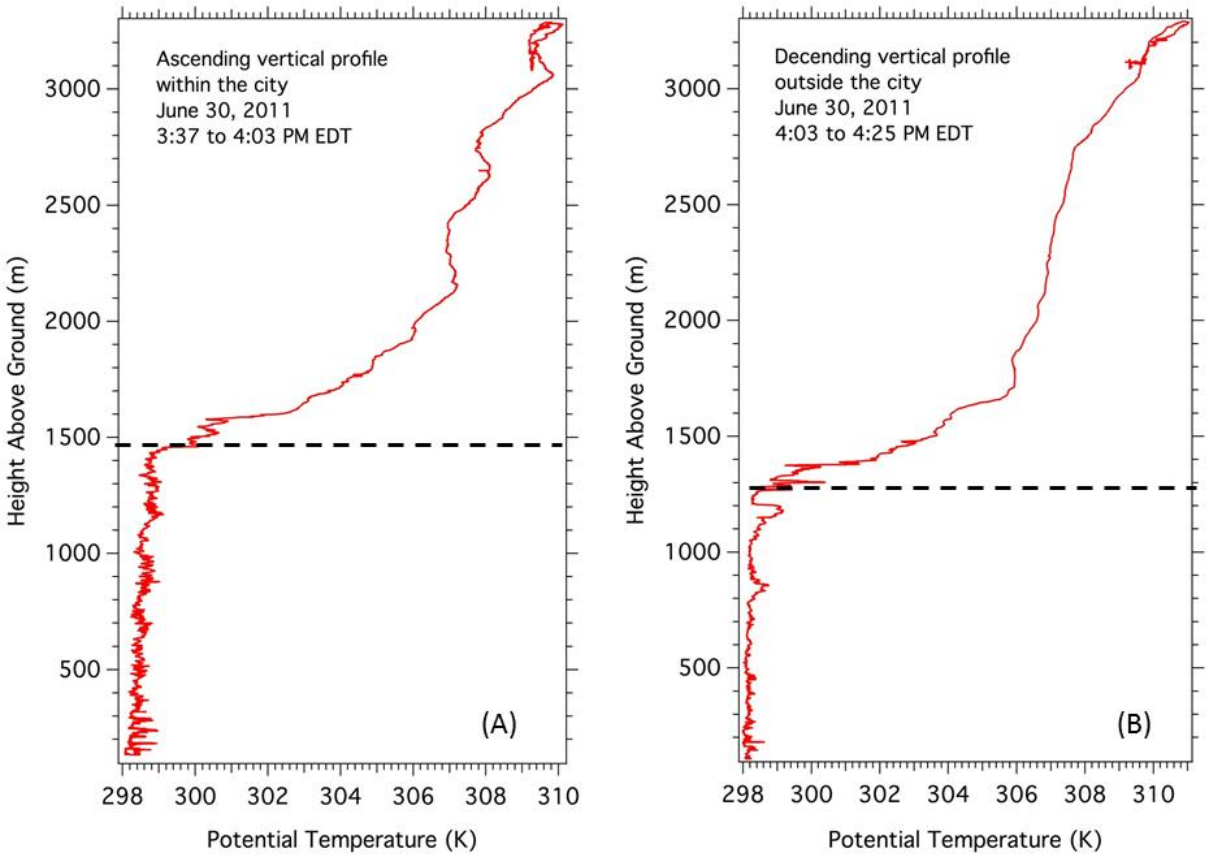
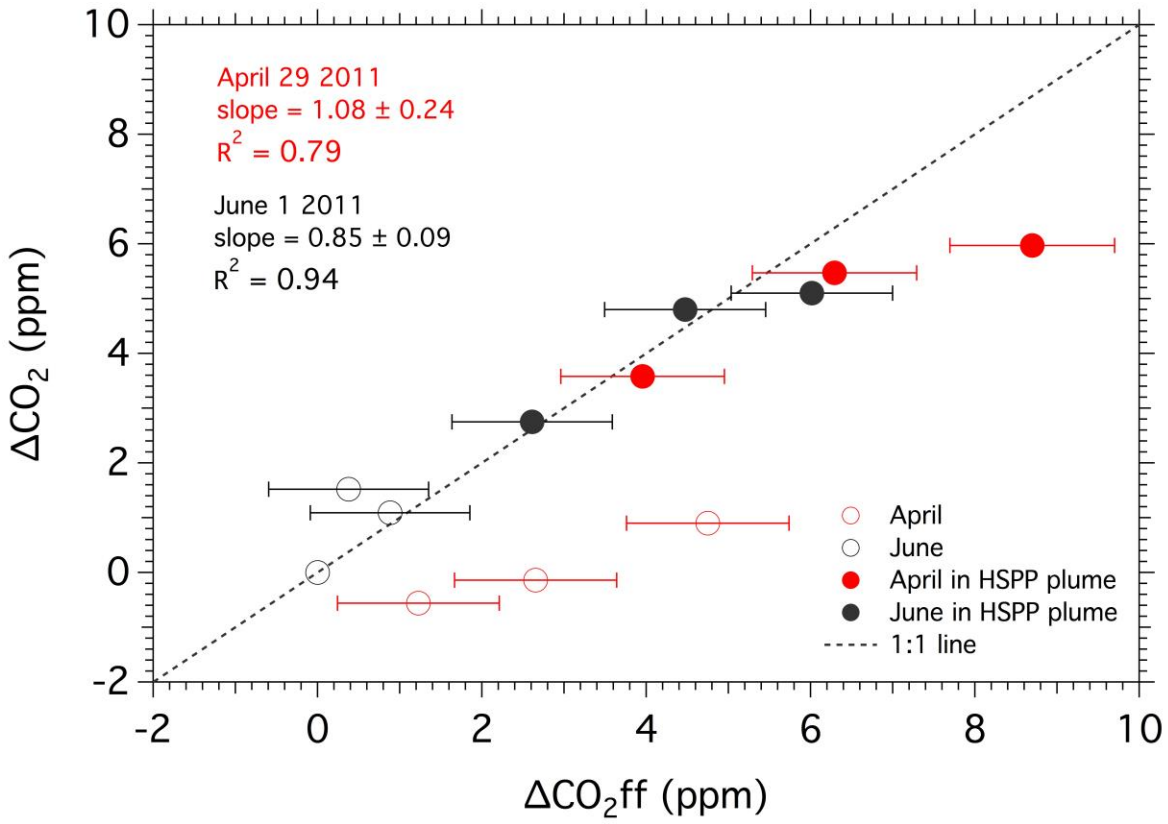


Figure S9. Aircraft vertical profiles of potential temperature conducted 30-minutes apart on 30 June 2011 above (A) an INFLUX tower site located inside the city of Indianapolis (urban site), and (B) a forested site in Moorseville, IN (rural site), which is located in southwest of Indianapolis.



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136 Figure S10. Total CO_2 versus fossil fuel CO_2 from flask measurements on 29 April and 01 June,
137 2011 flight experiments. Filled circles correspond to flask measurements sampled downwind of
138 the Harding Street Power Plant (HSPP) plume. Radiocarbon CO_2 was not measured on 01
139 March 2011.

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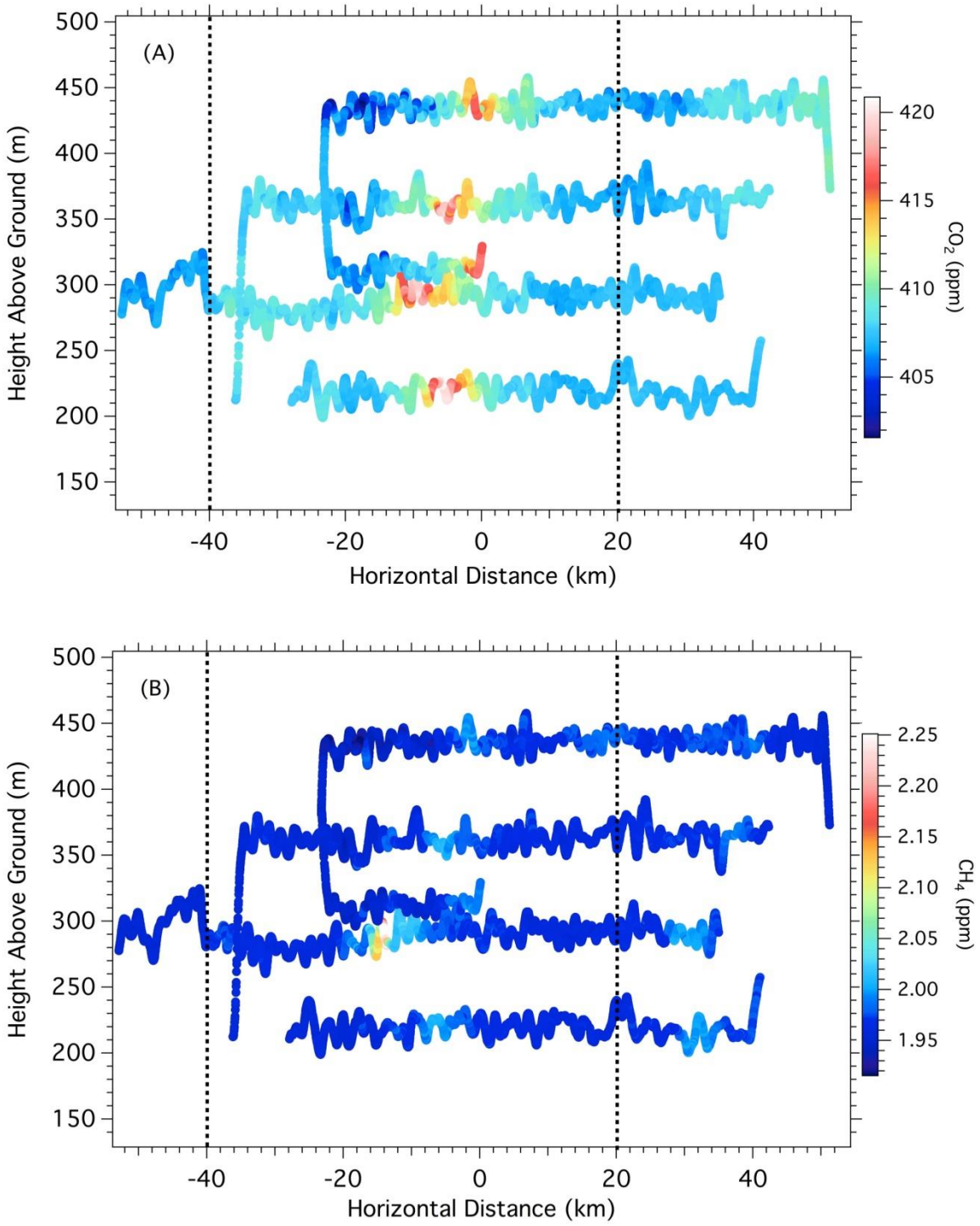

147 Figure S11. (A) CO_2 and (B) CH_4 observations along the horizontal transects for the various
148 altitudes on 01 March 2011. CBL depth was 525 m. Broken lines represent the minimum and
149 maximum horizontal limits of the city.