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Supporting Information for

Observing system choice can minimize interference of the biosphere in studies of urban CO_2 emissions

Raj M. Lal[1], Eric A. Kort[1]

[1]Climate and Space Sciences and Engineering, University of Michigan, Ann Arbor, MI., USA

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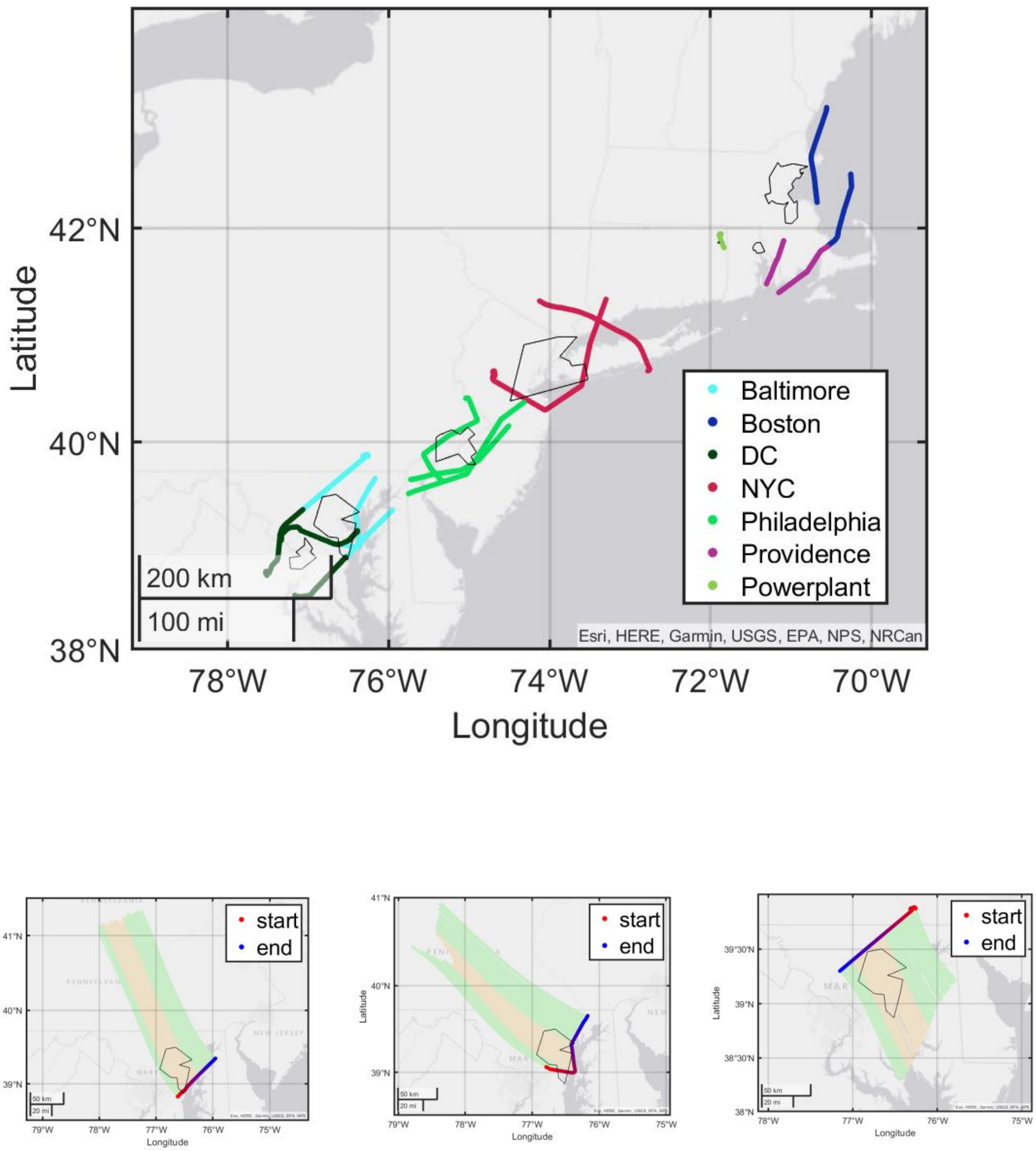
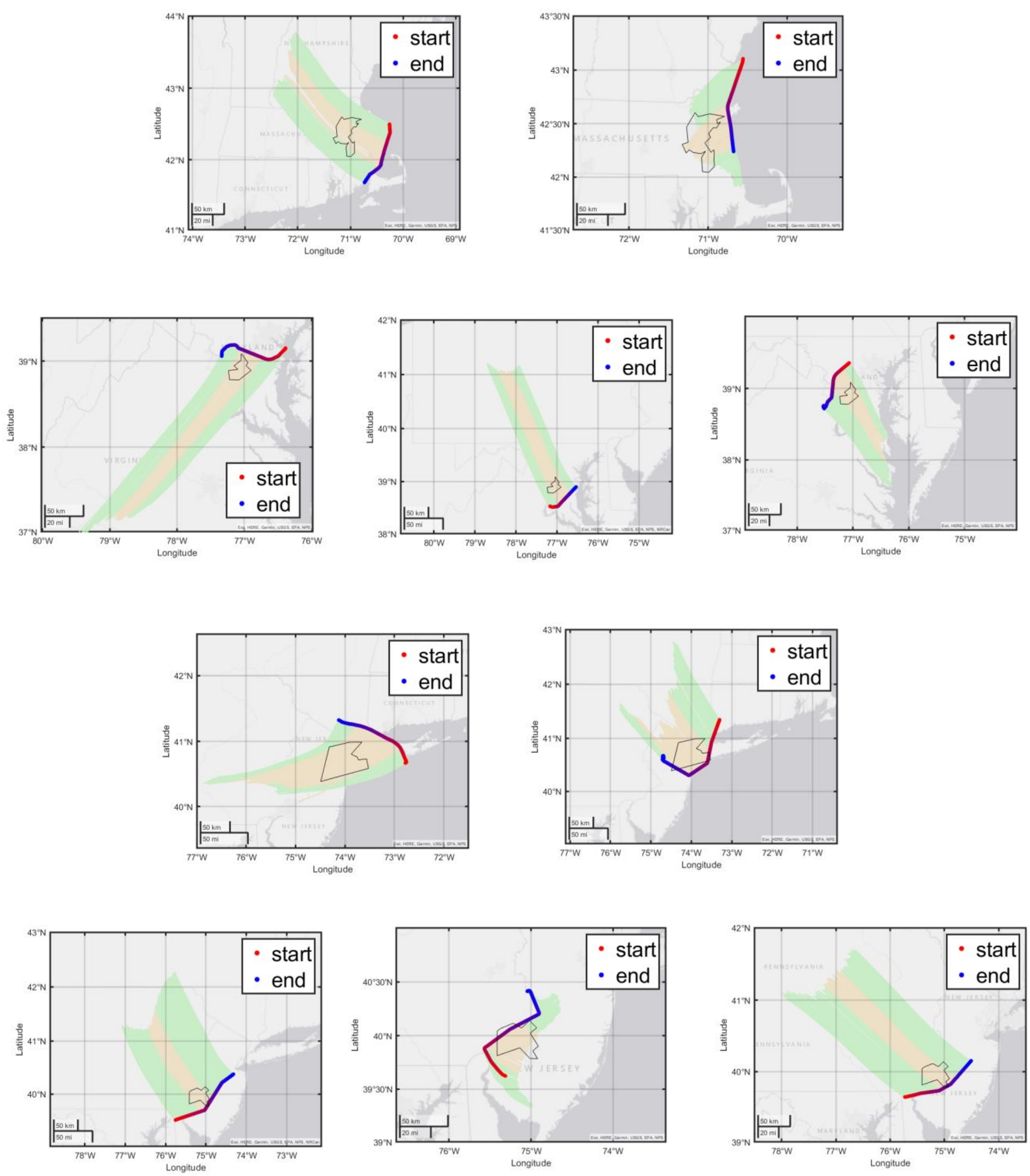
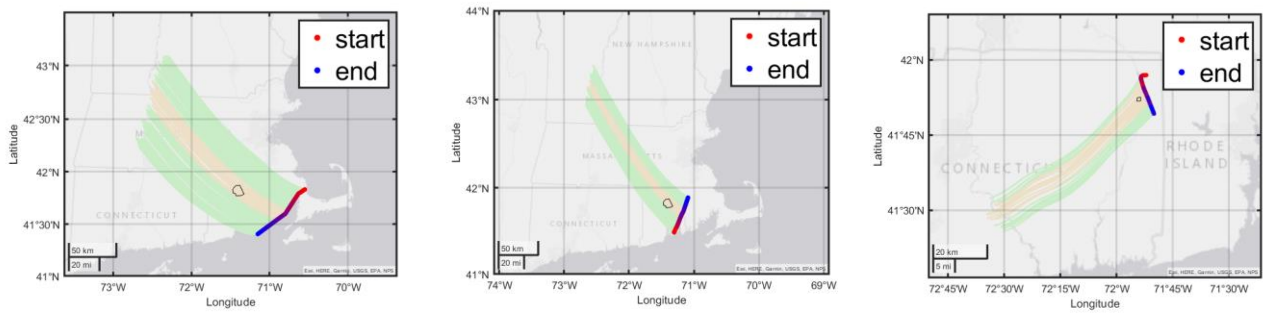
SI Section 1: Justifying a 6-hour Stochastic Time-Inverted Lagrangian Transport (STILT) back trajectory for characterizing urban CO_2 enhancements with a local background observing framework

Studies that assess CO_2 profiles from tower observations to understand urban CO_2 generally use air masses that have traveled ~days back in time. However, when assessing an urban core against its direct urban background with a local background observing framework, e.g., downwind airborne transects, where air masses upwind of the urban core are likely capturing similar surface influences, it is less necessary to assess upwind influences – assessed air masses need only to traverse the urban core. In addition, running STILT at fine resolutions 72-hours back in time can become prohibitively computationally expensive. Thus, we choose a time (6 hours) to run STILT backwards that can capture the entire urban air mass across cities while still capturing the regional background CO_2 contribution (see SI Fig. 1 confirming each urban area is captured with a 6-hour back trajectory). The focus of this observing framework is to measure the urban-suburban-rural gradient along a transect – what happens far upwind of the city is mostly homogenous and doesn't directly affect the local gradient. We use a base number of hours and not transect-dependent number of hours for computational efficiency purposes. Here, we utilize the NYC-0413 transect, as this was the only transect whose 72-hour back trajectory stayed within the Continental US (where we had surface emission flux data) (SI Fig. 3). We found that >90% of the CO_2 fossil fuel and combined (fossil fuel + biosphere) source contribution within each season along the airborne transect occurred within 6 hours of the ensemble being released compared to a 72-hour run (SI Fig. 6), suggesting the 6-hour approach adequately captures near-field impacts and can be used to assess urban CO_2 profiles that are assessed with a local background observing framework.

SI Fig. 1: The airborne transects whose urban CO_2 outflows were used in model space for this study, including modeled city boundaries and STILT-derived, 6-hour back trajectories of receptors along each downwind transect. In the transect-specific back-trajectory plots, the tan region indicates back trajectories along receptors that are defined as urban while the green swaths represent receptors that are defined as urban-background.




start
end
Latitude
Longitude
NEW HAMPSHIRE
MASSACHUSETTS
CONNECTICUT
VIRGINIA
NEW JERSEY
PENNSYLVANIA
MARYLAND
50 km
20 mi
50 mi


start
end
Latitude
Longitude
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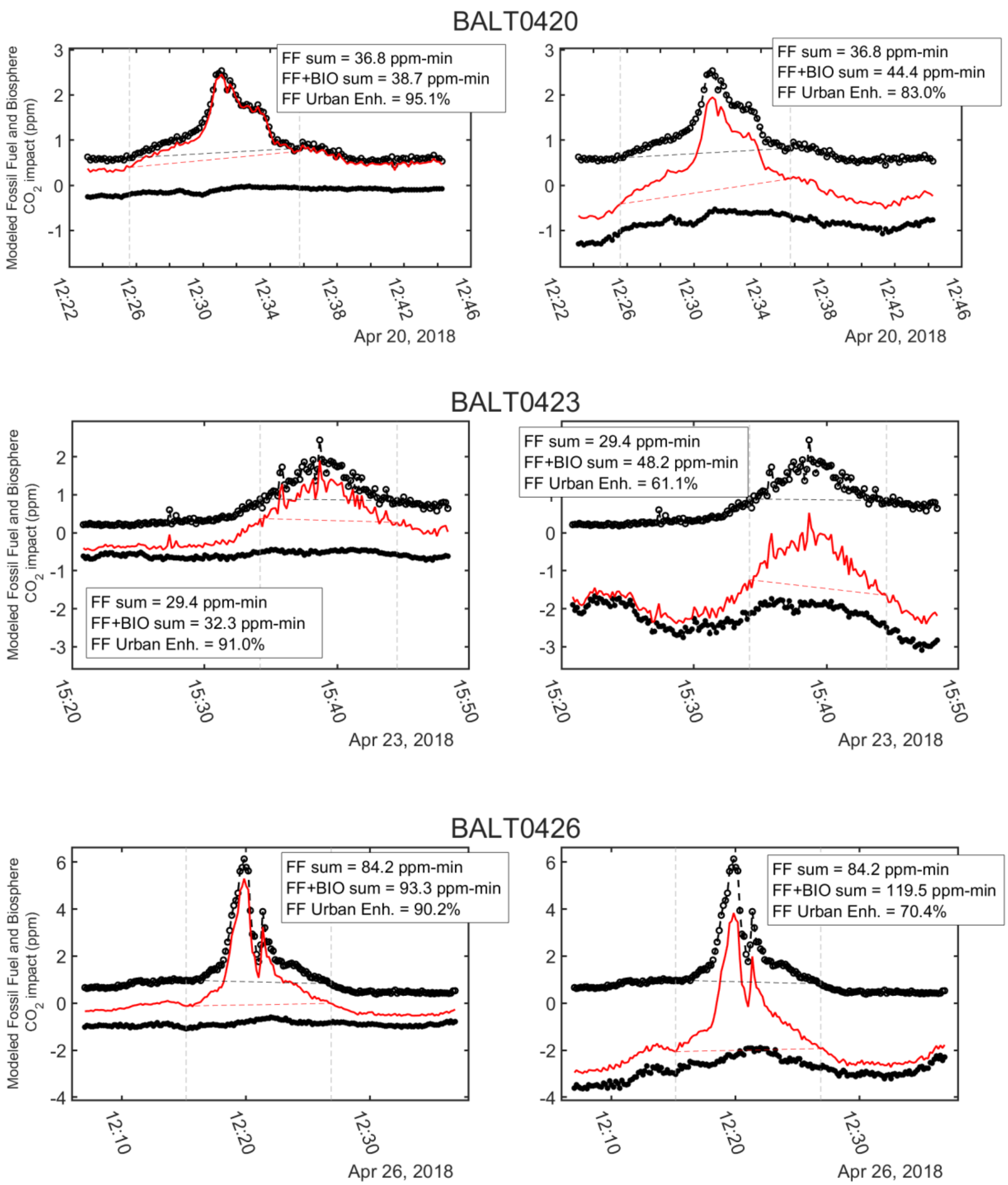
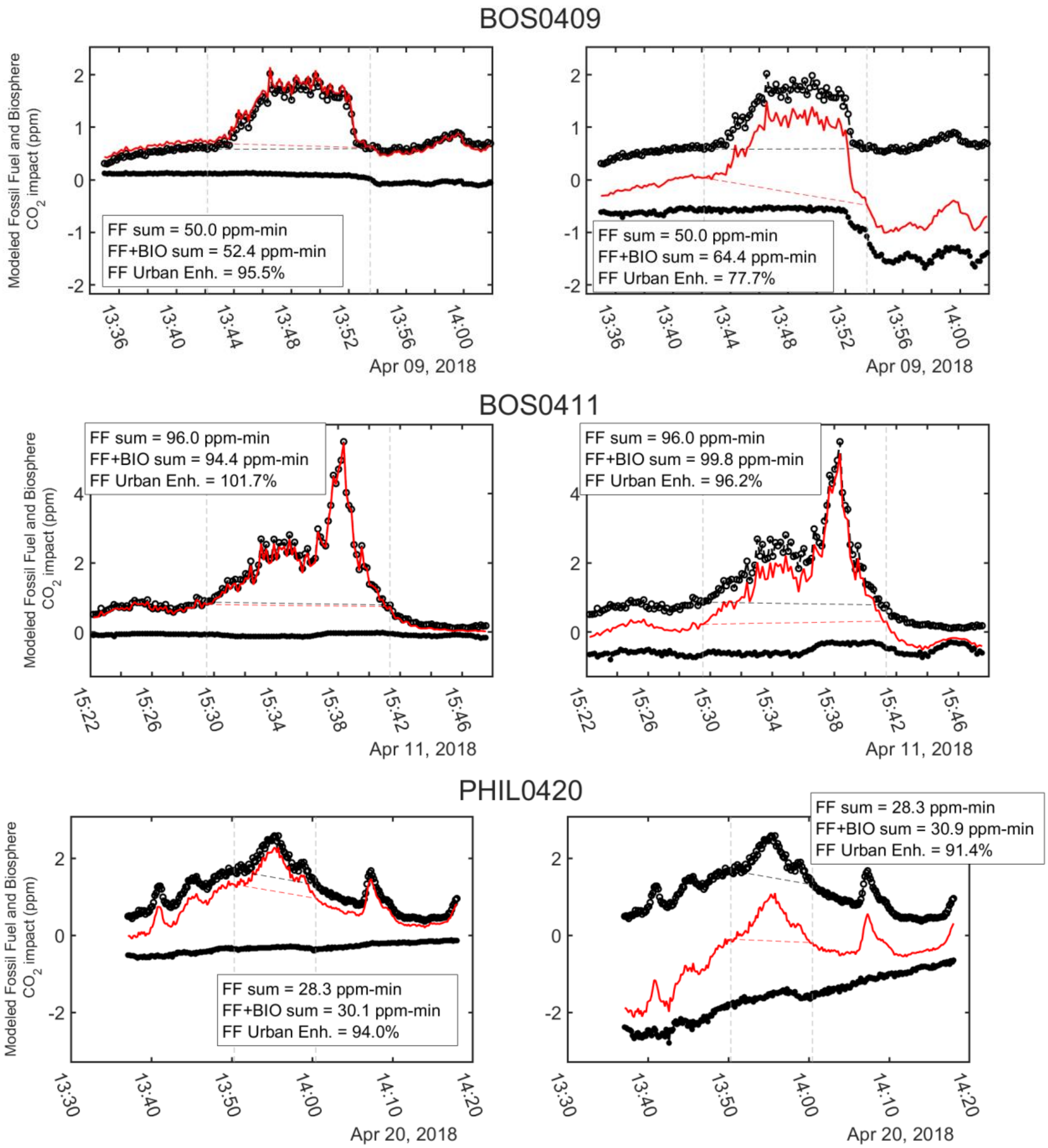
42°30'N
42°N
41°30'N
41°N
73°W
72°W
71°W
70°W
CONNECTICUT
50 km
20 mi
44°N
74°W
69°W
NEW HAMPSHIRE
42°N
41°45'N
41°30'N
72°45'W
72°30'W
72°15'W
71°45'W
71°30'W
RHODE
ISLAND
20 km
5 mi

SI Fig. 2: Fossil fuel and biosphere CO_2 source contributions on the day of each airborne transect for each transect assessed in this study, shown with both modeled biosphere representations: (left column) VPRM and (right column) SMUrF. In each plot, the fossil fuel contribution is indicated by the black lines with open circles, the biosphere representation is indicated by the black lines with closed circles, and the combined (fossil fuel plus biosphere) impact is indicated by the red line.





BOS0409
Modeled Fossil Fuel and Biosphere CO_2 impact (ppm)
FF sum = 50.0 ppm-min
FF+BIO sum = 52.4 ppm-min
FF Urban Enh. = 95.5%
FF sum = 50.0 ppm-min
FF+BIO sum = 64.4 ppm-min
FF Urban Enh. = 77.7%
13:36
13:40
13:44
13:48
13:52
13:56
14:00
Apr 09, 2018
BOS0411
FF sum = 96.0 ppm-min
FF+BIO sum = 94.4 ppm-min
FF Urban Enh. = 101.7%
FF sum = 96.0 ppm-min
FF+BIO sum = 99.8 ppm-min
FF Urban Enh. = 96.2%
15:22
15:26
15:30
15:34
15:38
15:42
15:46
Apr 11, 2018
PHIL0420
FF sum = 28.3 ppm-min
FF+BIO sum = 30.1 ppm-min
FF Urban Enh. = 94.0%
FF sum = 28.3 ppm-min
FF+BIO sum = 30.9 ppm-min
FF Urban Enh. = 91.4%
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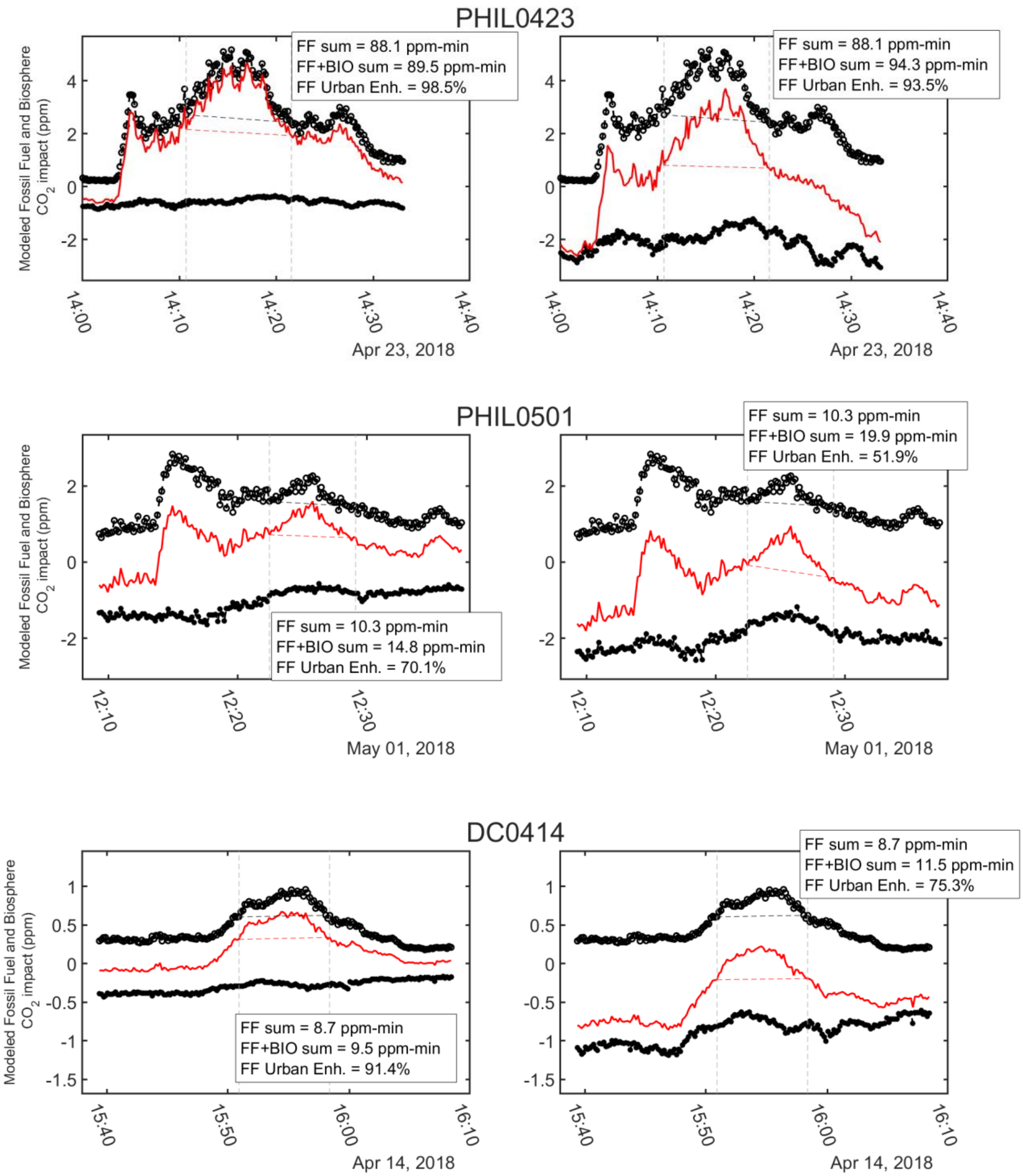
13:40
13:50
14:00
14:10
14:20
Apr 20, 2018


PHIL0423
Modeled Fossil Fuel and Biosphere CO2 impact (ppm)
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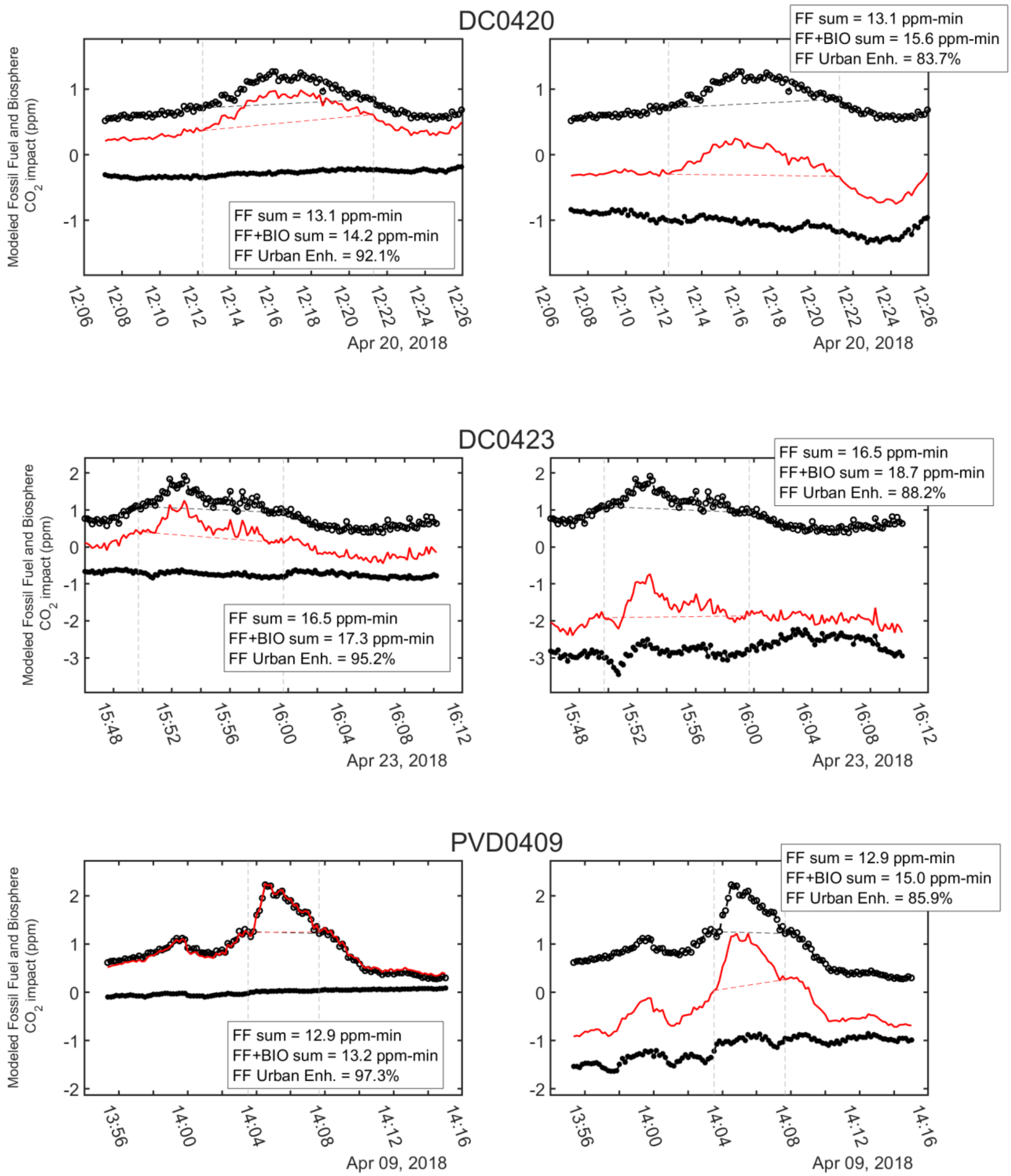
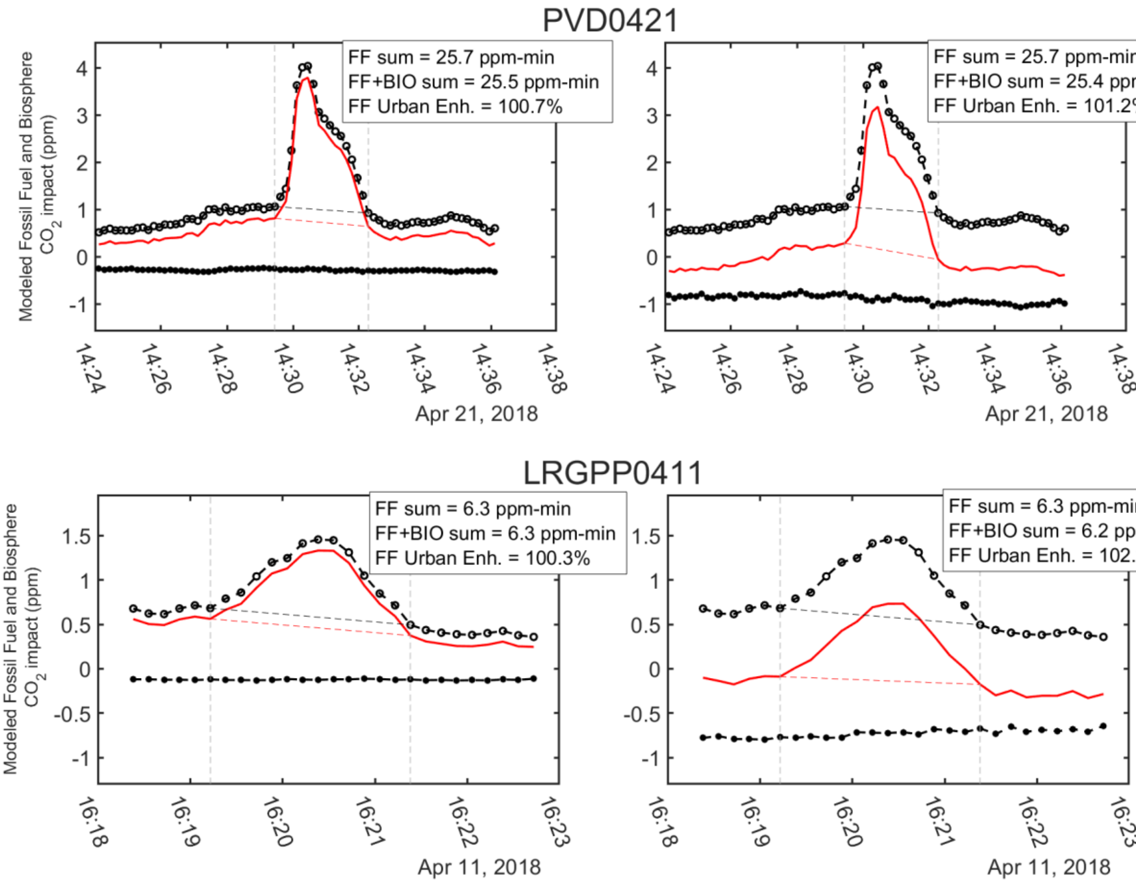
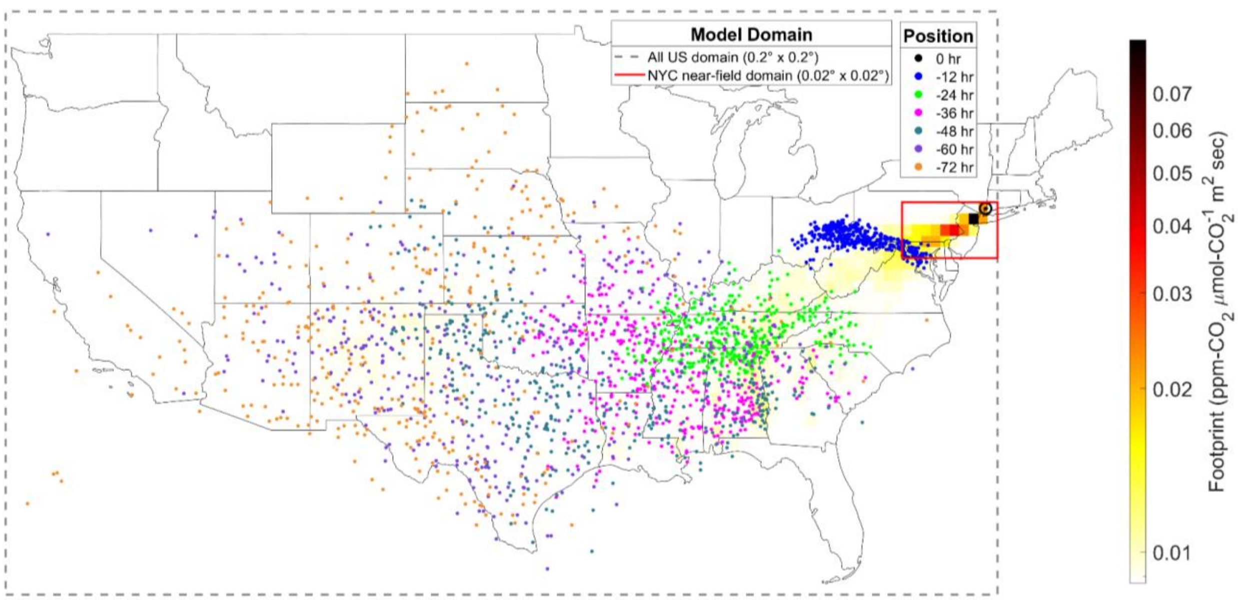
FF+BIO sum = 89.5 ppm-min
FF Urban Enh. = 98.5%
FF sum = 88.1 ppm-min
FF+BIO sum = 94.3 ppm-min
FF Urban Enh. = 93.5%
14:00
14:10
14:20
14:30
14:40
Apr 23, 2018
PHIL0501
FF sum = 10.3 ppm-min
FF+BIO sum = 14.8 ppm-min
FF Urban Enh. = 70.1%
FF sum = 10.3 ppm-min
FF+BIO sum = 19.9 ppm-min
FF Urban Enh. = 51.9%
12:10
12:20
12:30
May 01, 2018
DC0414
FF sum = 8.7 ppm-min
FF+BIO sum = 9.5 ppm-min
FF Urban Enh. = 91.4%
FF sum = 8.7 ppm-min
FF+BIO sum = 11.5 ppm-min
FF Urban Enh. = 75.3%
15:40
15:50
16:00
16:10
Apr 14, 2018


DC0420
FF sum = 13.1 ppm-min
FF+BIO sum = 14.2 ppm-min
FF Urban Enh. = 92.1%
FF sum = 13.1 ppm-min
FF+BIO sum = 15.6 ppm-min
FF Urban Enh. = 83.7%
Modeled Fossil Fuel and Biosphere
CO2 impact (ppm)
Apr 20, 2018
DC0423
FF sum = 16.5 ppm-min
FF+BIO sum = 17.3 ppm-min
FF Urban Enh. = 95.2%
FF sum = 16.5 ppm-min
FF+BIO sum = 18.7 ppm-min
FF Urban Enh. = 88.2%
Apr 23, 2018
PVD0409
FF sum = 12.9 ppm-min
FF+BIO sum = 13.2 ppm-min
FF Urban Enh. = 97.3%
FF sum = 12.9 ppm-min
FF+BIO sum = 15.0 ppm-min
FF Urban Enh. = 85.9%
Apr 09, 2018

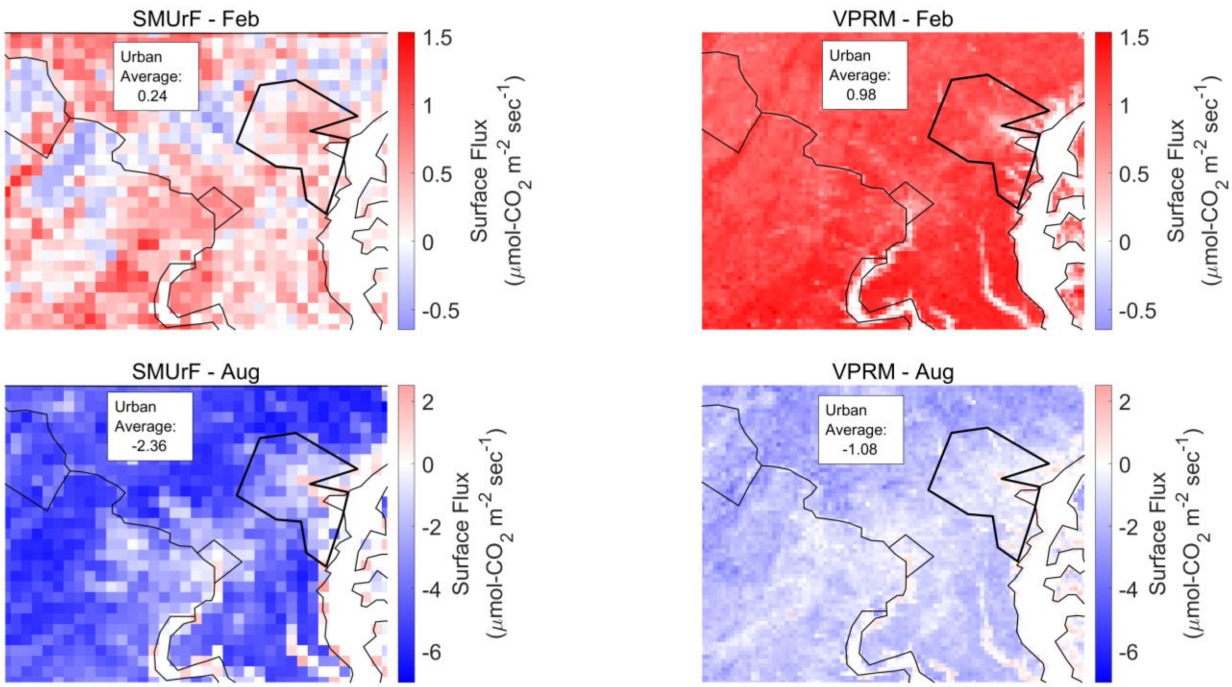

PVD0421
FF sum = 25.7 ppm-min
FF+BIO sum = 25.5 ppm-min
FF Urban Enh. = 100.7%
FF sum = 25.7 ppm-min
FF+BIO sum = 25.4 ppm-min
FF Urban Enh. = 101.2%
Modeled Fossil Fuel and Biosphere CO2 impact (ppm)
Apr 21, 2018
LRGPP0411
FF sum = 6.3 ppm-min
FF+BIO sum = 6.3 ppm-min
FF Urban Enh. = 100.3%
FF sum = 6.3 ppm-min
FF+BIO sum = 6.2 ppm-min
FF Urban Enh. = 102.2%
Apr 11, 2018

SI Fig. 3: Location of 400 released air parcels and surface footprint profile from an idealized-located tower site downwind of New York City as part of the NYC-0413 transect (the only transect whose 72-hour back trajectory stayed in CONUS). The 72-hour source contribution approach included the near-field domain (first 6 hours) where STILT was run at a 0.02°x0.02° resolution and the far-field domain where STILT was run at a 0.2°x0.2° resolution

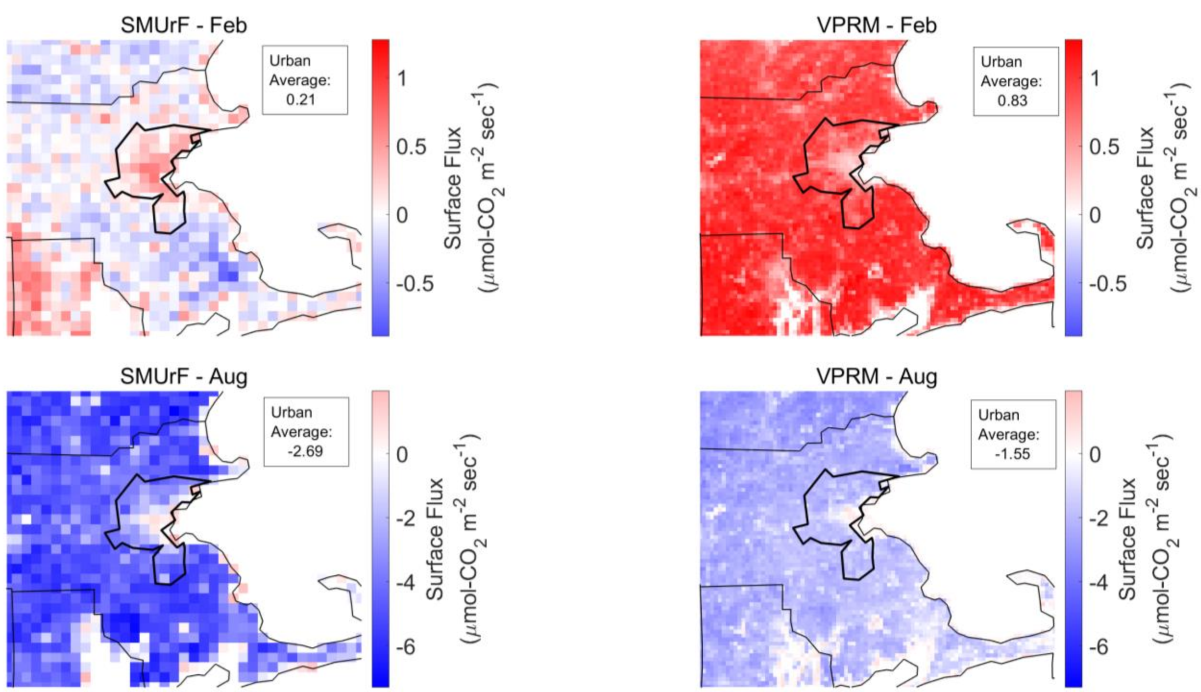


SI Fig. 4: Gridded SMUrF and VPRM surface fluxes for a winter and summer month (February and August, respectively) across urban study domains

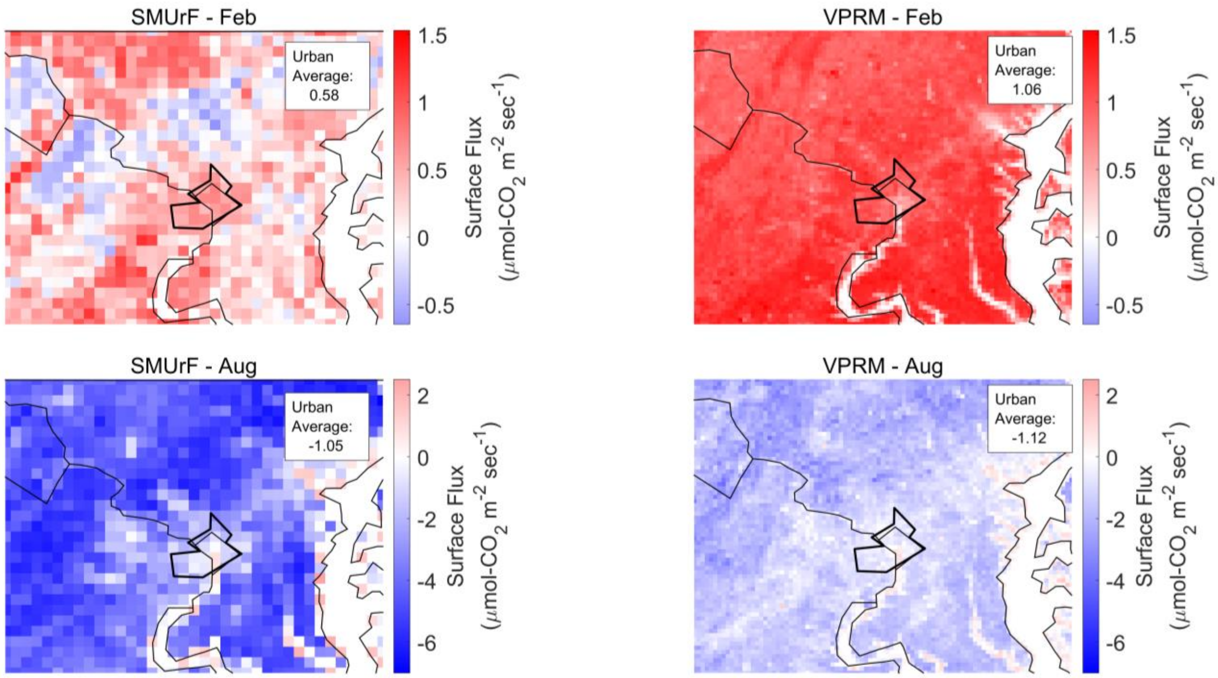
BALTIMORE



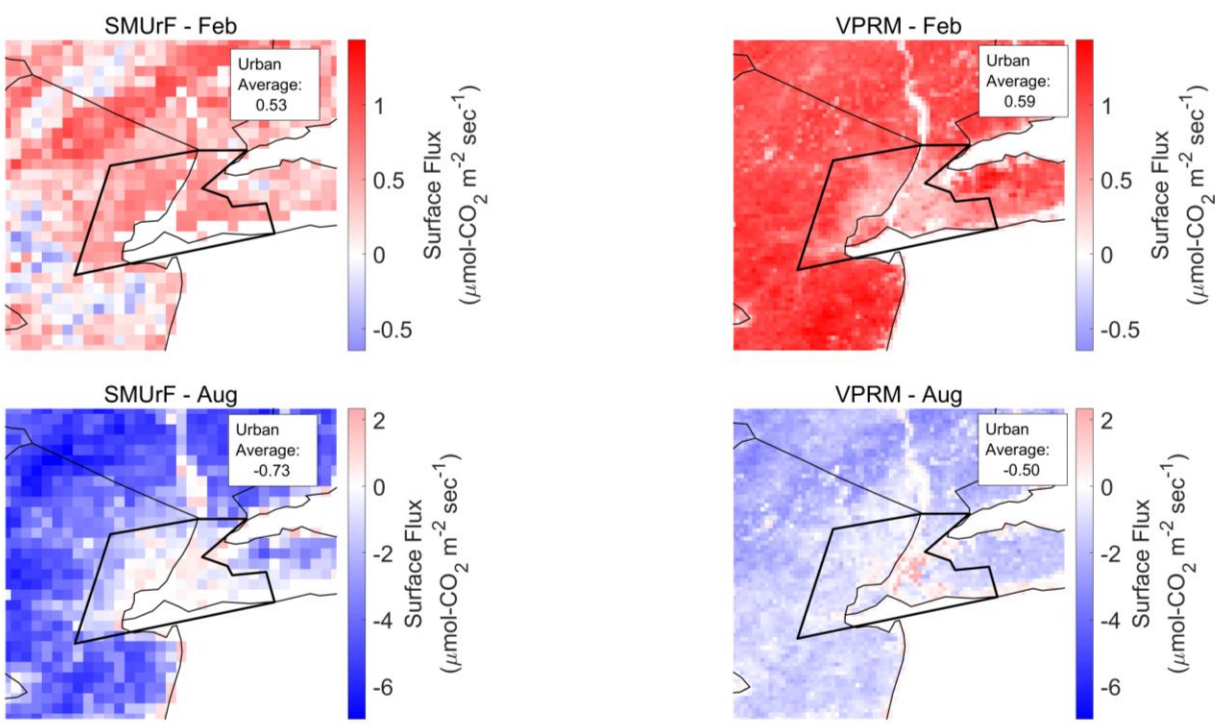
BOSTON



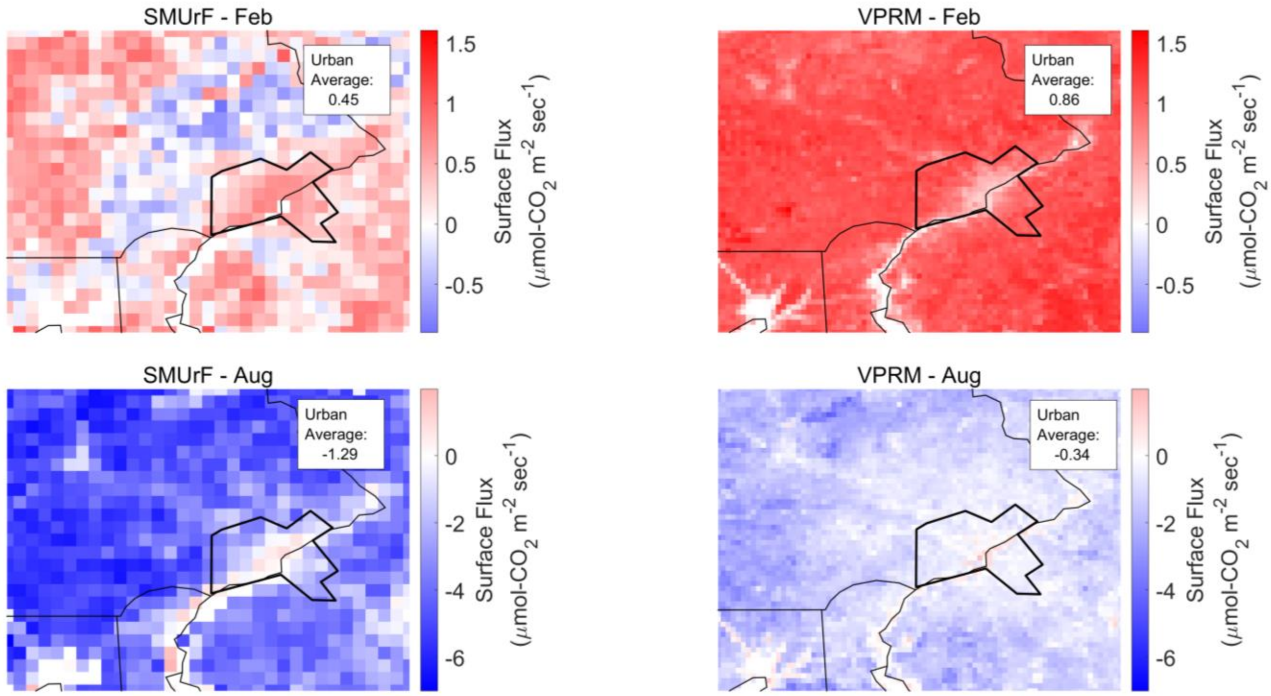
WASHINGTON D.C.



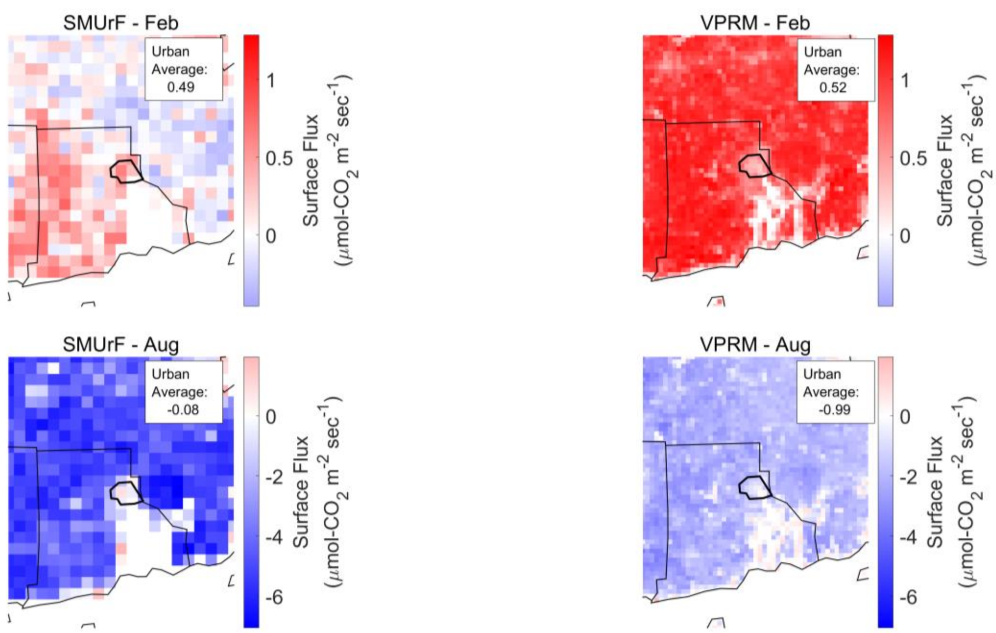
NEW YORK CITY



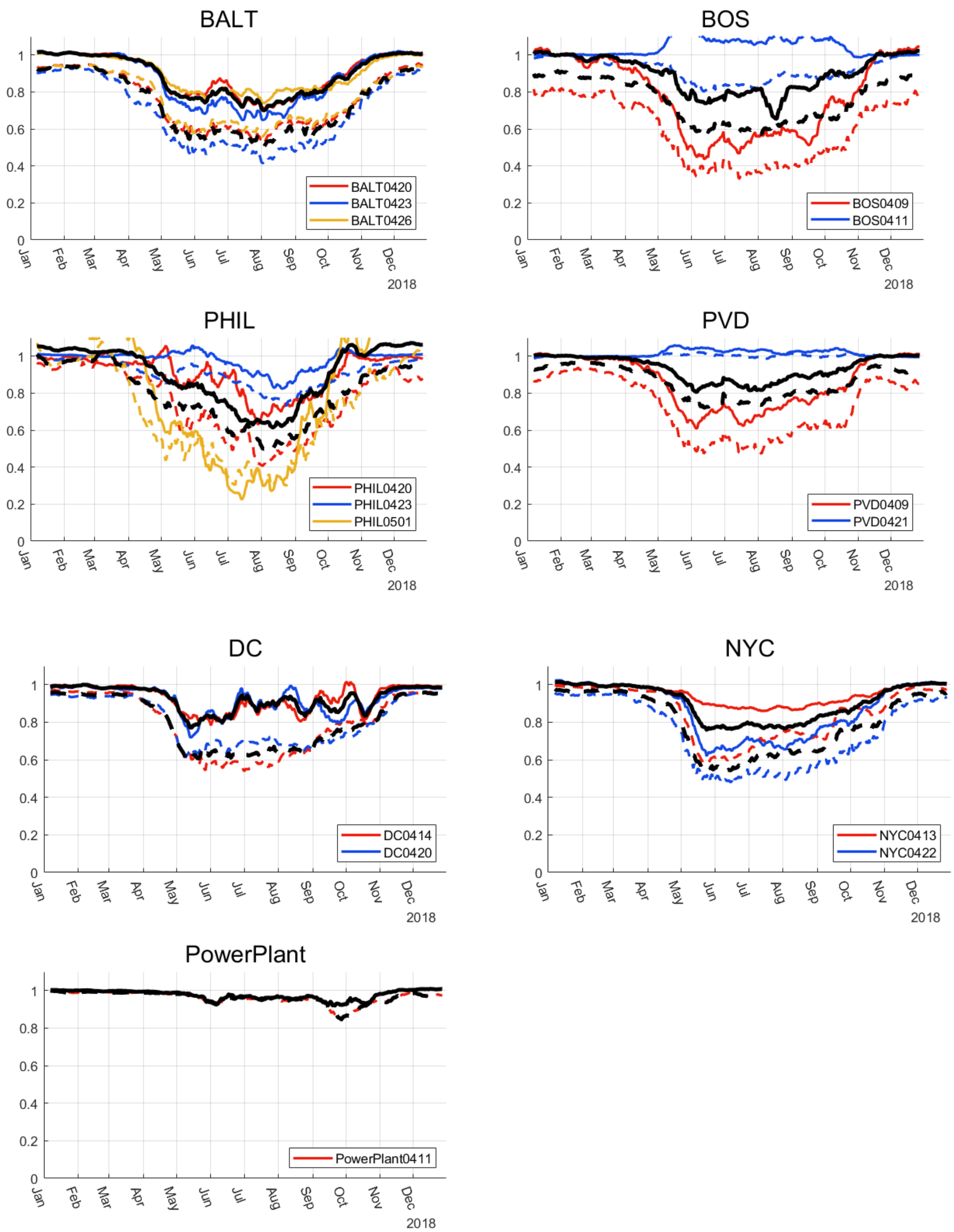
PHILADELPHIA



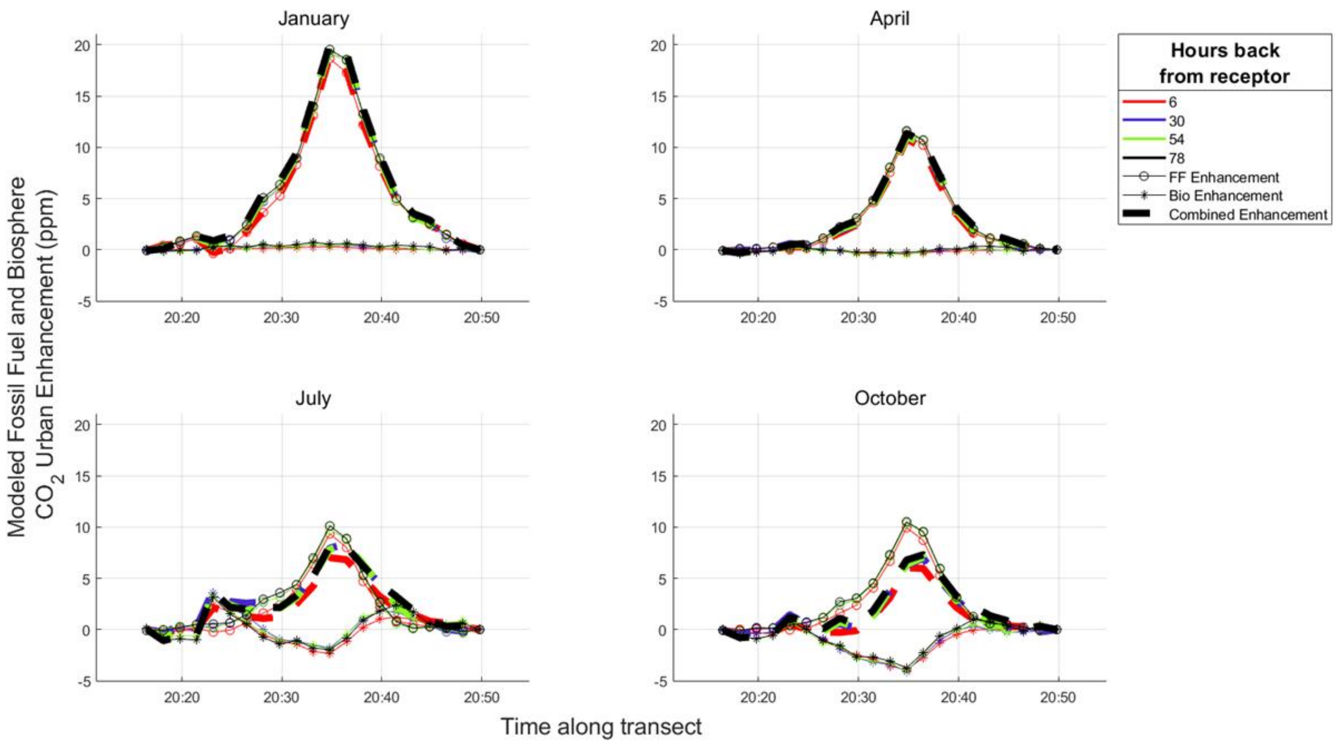
PROVIDENCE



SI Fig. 5: Simulated fossil fuel contribution (%) for each airborne transect in each study domain (6 cities and 1 power plant) over the course of a year. The averages of each city are shown in Main Text Fig. 2.



SI Fig. 6: One-week averaged fossil fuel, biosphere, and combined (fossil fuel + biosphere) CO_2 enhancement across seasons for the NYC-0413 transect modeled 78 hours back in time. We find that across seasons, the first 6 hours captures >90% of the combined CO_2 enhancement.



SI Table 1: Comparison of percent differences between the two calculation approaches as shown in Main Text Eqn. 1 across all urban areas for all months. The highlighted value indicates the percent difference that was shown in Main Text Table 2 and used in the analysis

Month	City	Average Urban Surface Flux ($nmol\ m^{-2} s^{-1}$) VPRM	Average Urban Surface Flux ($nmol\ m^{-2} s^{-1}$) SMUrF	Percent Difference Eqn. 1A	Percent Difference Eqn. 1B
JAN	BALT	772	6	197	99
	BOS	627	102	144	84
	DC	699	301	80	57
	NYC	422	380	11	10
	PHIL	709	266	91	62
	PVD	363	326	11	10
	All City Average	***599***	***230***	***89***	***54***
FEB	BALT	980	237	122	76
	BOS	831	206	121	75
	DC	1058	583	58	45
	NYC	592	526	12	11
	PHIL	861	445	64	48
	PVD	516	486	2	6
	All City Average	***806***	***414***	***63***	***44***
MAR	BALT	817	-87	247	111
	BOS	547	170	105	69
	DC	783	186	123	76
	NYC	458	344	28	25
	PHIL	751	168	127	78
	PVD	402	303	28	25
	All City Average	***626***	***181***	***110***	***64***
APR	BALT	634	-366	747	158
	BOS	546	-254	547	146
	DC	606	-9	206	101
	NYC	444	240	60	46
	PHIL	530	-83	274	116
	PVD	401	168	82	58
	All City Average	***527***	***-51***	***319***	***104***
MAY	BALT	-1195	-2897	83	59
	BOS	-1984	-2669	29	26
	DC	-1089	-1660	42	34
	NYC	-443	-865	65	49
	PHIL	-653	-1442	75	55
	PVD	-1058	-463	78	56
	All City Average	***-1070***	***-1666***	***62***	***46***
JUNE	BALT	-2148	-4710	75	54
	BOS	-4259	-5853	32	27
	DC	-1646	-2814	52	41
	NYC	-1487	-1902	25	22
	PHIL	-1874	-3059	48	39
	PVD	-2640	-1283	69	51
	All City Average	***-2342***	***-3270***	***50***	***39***

Month	City	Average Urban Surface Flux ($nmol\ m^{-2} s^{-1}$) VPRM	Average Urban Surface Flux ($nmol\ m^{-2} s^{-1}$) SMUrF	Percent Difference Eqn. 1A	Percent Difference Eqn. 1B
JUL	BALT	-1864	-3427	59	46
	BOS	-3354	-4268	24	21
	DC	-1349	-1957	37	31
	NYC	-995	-1667	50	40
	PHIL	-1463	-2474	51	41
	PVD	-2338	-685	109	71
	All City Average	***-1894***	***-2413***	***55***	***42***
AUG	BALT	-1082	-2360	74	54
	BOS	-1552	-2691	54	42
	DC	-1122	-1048	7	7
	NYC	-504	-726	36	31
	PHIL	-342	-1293	116	74
	PVD	-993	-82	169	92
	All City Average	***-933***	***-1367***	***76***	***50***
SEP	BALT	1219	-772	891	163
	BOS	-517	-1837	112	72
	DC	1250	-21	207	102
	NYC	723	-255	418	135
	PHIL	1464	-364	333	125
	PVD	-133	106	1764	180
	All City Average	***668***	***-524***	***621***	***129***
OCT	BALT	1173	-534	535	146
	BOS	593	-812	1278	173
	DC	1122	-7	203	101
	NYC	674	40	178	94
	PHIL	766	-111	268	115
	PVD	381	224	52	41
	All City Average	***785***	***-200***	***419***	***112***
NOV	BALT	1049	-31	212	103
	BOS	918	128	151	86
	DC	973	279	111	71
	NYC	683	415	49	39
	PHIL	940	207	128	78
	PVD	678	349	64	48
	All City Average	***874***	***225***	***119***	***71***
DEC	BALT	956	549	54	43
	BOS	759	495	42	35
	DC	862	759	13	12
	NYC	577	692	18	17
	PHIL	846	709	18	16
	PVD	513	561	9	9
	All City Average	***752***	***627***	***26***	***22***

SI Table 2: Monthly fossil fuel contribution (% of simulated CO_2 enhancement) within each of the study domains (6 cities and 1 power plant) for both biosphere representations (VPRM and SMUrF). The monthly averages presented here are the averages of the 6-hour daily period aligned with the STILT simulations across model spaces

	All Site Average		BALTIMORE		BOSTON		DC		NYC		PHILADELPHIA		PROVIDENCE		POWER PLANT	
	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF	VPRM	SMURF
Jan	1.02	0.95	1.01	0.93	1.01	0.91	1.00	0.95	1.01	0.96	1.04	0.97	1.04	0.96	1.04	0.99
Feb	1.01	0.95	1.01	0.94	0.99	0.90	0.99	0.94	1.00	0.96	1.04	0.98	1.04	0.97	1.04	0.99
Mar	1.01	0.94	0.99	0.90	0.99	0.88	0.98	0.93	0.99	0.94	1.03	0.99	1.03	0.97	1.04	0.99
Apr	0.96	0.84	0.96	0.80	0.95	0.85	0.96	0.85	0.97	0.88	0.97	0.82	0.96	0.84	0.98	0.89
May	0.83	0.68	0.80	0.60	0.85	0.68	0.82	0.63	0.83	0.62	0.85	0.74	0.81	0.73	0.88	0.86
Jun	0.78	0.63	0.78	0.57	0.77	0.60	0.83	0.62	0.78	0.58	0.79	0.68	0.72	0.66	0.82	0.81
Jul	0.75	0.61	0.75	0.56	0.80	0.61	0.87	0.63	0.79	0.62	0.67	0.58	0.66	0.63	0.75	0.78
Aug	0.74	0.61	0.74	0.56	0.78	0.62	0.85	0.64	0.79	0.63	0.64	0.53	0.69	0.63	0.77	0.77
Sep	0.82	0.68	0.79	0.60	0.82	0.65	0.86	0.71	0.84	0.69	0.79	0.66	0.82	0.73	0.88	0.83
Oct	0.93	0.76	0.89	0.68	0.89	0.70	0.89	0.76	0.89	0.76	1.00	0.79	0.96	0.80	1.00	0.89
Nov	1.01	0.89	0.99	0.84	0.97	0.84	0.98	0.90	0.98	0.89	1.04	0.92	1.04	0.93	1.05	0.96
Dec	1.03	0.94	1.01	0.93	1.01	0.90	0.99	0.95	1.00	0.94	1.06	0.95	1.06	0.95	1.06	0.99