TITLE

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

AUTHORS

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KEYWORDS

Urban CO_2, Biosphere, Fossil Fuel Emissions, Greenhouse Gas Observations

PLAIN LANGUAGE SUMMARY

Cities around the world have announced plans to reduce CO_2 emissions. Atmospheric CO_2 observations provide a potential pathway toward independent assessment of implemented policies. However, these measurements can be strongly influenced by the urban biosphere, which can act as both a source (respiration) and sink (photosynthesis) of CO_2. If using an observing approach that introduces a local, urban background – e.g., observations via a downwind airborne transect that captures an entire urban outflow – the relative role of the biosphere may be minimized. Here, we combine back trajectory modeling with high-resolution surface fossil fuel and biosphere CO_2 fluxes across six cities and one powerplant in the NE US to demonstrate that observing strategies using this approach can greatly reduce biosphere interferences in studies of urban CO_2 (<10% biosphere interference outside of summer months, on average) and pave the way to conduct robust studies of urban fossil fuel CO_2 emissions.

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ABSTRACT

Cities around the world have introduced initiatives to reduce CO_2 emissions. Atmospheric observations can provide evaluation and assessment of these initiatives by quantifying emissions, considering local sources and sinks. The relative importance of the urban biosphere, which can act as both a source (respiration) and sink (photosynthesis) of CO_2, has previously been suggested to strongly impact urban CO_2 measurements, confounding the ability to use observations to study fossil emissions. However, if using an observing framework that measures a local urban background and the direct urban core outflow, e.g., along a downwind airborne transect, the biosphere's role may be minimized. Here, we combine real, airborne observations of CO_2 downwind of select cities in the Northeast US with high-resolution, back-trajectory modeling and spatially-and temporally-resolved surface biosphere and fossil fuel fluxes to characterize the relative biosphere importance to urban CO_2 profiles. We show the biosphere influence using this urban observing system to be small, averaging only 15% of the local CO_2 enhancement annually, <10% outside of summer, with a maximum influence of 29% in summer when the biosphere drawdown is most pronounced. Furthermore, when considering two biosphere models that differ by >80% , the impact on observed urban CO_2 signals is reduced to only 12% on average. Urban observing frameworks that utilize this local background approach – including those via aircraft or satellite observations – can minimize the biosphere's influence and thus help facilitate robust assessments of urban fossil fuel CO_2 emissions.

INTRODUCTION

Cities across the world have announced initiatives to reduce anthropogenic greenhouse gas (GHG) emissions to combat climate change [1, 2]. CO_2 has been identified as the main GHG of concern as it is the primary GHG emitted through human activities and is the single largest driver of our changing climate. In the United States, urban areas contribute ~40% of nationwide CO_2 emissions [3, 4], so studying and assessing CO_2 profiles in urban areas, particularly their sources and sinks, can be critical to address climate concerns. Specifically, it can: 1) help inform climate policy by providing direct information on emissions and assessing whether implemented policies are having desired effects and 2) support fundamental carbon cycle research by constraining human inputs into the atmosphere to disentangle changes driven by anthropogenic emissions or biosphere dynamics.

The biosphere acts both as a source (via respiration) and a sink (via photosynthesis) of CO_2, and because of its potential significance to understanding urban CO_2, a number of previous studies have emphasized the biosphere and the role it plays. Previous representations of urban domains, including via isotopic measurements [5] and tower observations coupled with emission inventories [6] have suggested the biosphere is a substantial contributor to observed CO_2 in urban domains [7-9]. These results have particular pertinence for the expanding networks of urban tower observations in the US, including Los Angeles [10, 11], the Northeast Corridor [12], Indianapolis [13], Boston [6], and Salt Lake City [14]. However, if an urban area is characterized with an observing strategy where, at the city-scale, the urban signal can be isolated using measurements of the rural-suburban-urban boundary to define a local background (e.g., via an airborne platform that transects the full rural-suburban-urban outflow, or a similar observing strategy based on satellite-based remote sensing), the influence of the biosphere and the importance of how well it needs to be represented may not be as significant.

Here, we present a framework that utilizes real, airborne observations in model space to characterize the relative biosphere contribution to urban CO_2 enhancements. We apply this framework to select cities and a large point source in the Northeast U.S. over the course of a year to understand seasonal variation in biosphere contributions to CO_2 enhancements when assessed with a local background observing strategy for both whole city and large point source emission profiles. We use multiple biosphere representations to evaluate how different methods to estimate biosphere fluxes influences its relative significance to urban CO_2 observed with a local background framework. In addition, we compare the biosphere impacts estimated from this approach to what the biosphere contribution would be from an idealized-located tower downwind of one of the cities analyzed here. Lastly, we assess surface fluxes of fossil fuel and biosphere models/inventories within each of the urban cores studied here as a first-order, bottom-up comparison of the biosphere influence to urban CO_2 profiles.

METHODS

To ultimately understand the relative influence of the biosphere to an urban CO_2 enhancement observed with a local background observing framework approach (i.e., with an observing strategy where, at the city-scale, the urban signal can be isolated using measurements of the rural-suburban-urban-suburban-rural boundary to define a local background; e.g., via downwind airborne transects that capture the urban outflow), we use previously identified CO_2 outflows downwind of urban areas that were part of the East Coast Outflow 2018 (ECO-2018) campaign (New York City, NY; Boston, MA.; Philadelphia, PA.; Washington DC; Baltimore, MD.; Providence, RI.) [15]. The focus of this work is on urban CO_2 profiles, but we additionally apply this framework to one, large point source (Lake Road Generating Power Plant, VT.) to evaluate this approach's viability for point source assessments. These airborne transects (Table 1; SI Fig. 1), which were designed specifically to target urban emissions, were used as a basis for the modeling effort described next.

Table 1: The locations, dates, and times of CO_2 outflows identified as part of the ECO-2018 flight campaign. Observations along each of these flight paths were used in model space to assess the relative biosphere influence to urban/large point source CO_2 enhancements.

Model-Space Domain	Flight Date	Local Flight Time	Shorthand ID
Baltimore, MD	20-Apr-18	12:23-12:44	BALT-0420
Baltimore, MD	23-Apr-18	15:20-15:48	BALT-0423
Baltimore, MD	26-Apr-18	12:07-12:36	BALT-0426
Boston, MA	9-Apr-18	13:35-14:01	BOS-0409
Boston, MA	11-Apr-18	15:22-15:47	BOS-0411
Washington DC	14-Apr-18	15:39-16:08	DC-0414
Washington DC	20-Apr-18	12:07-12:25	DC-0420
New York City, NY	13-Apr-18	16:16-16:50	NYC-0413
New York City, NY	22-Apr-18	14:11-15:09	NYC-0422
Philadelphia, PA	20-Apr-18	13:37-14:17	PHIL-0420
Philadelphia, PA	23-Apr-18	14:00-14:33	PHIL-0423
Philadelphia, PA	1-May-18	12:09-12:37	PHIL-0501
Providence, RI	9-Apr-18	13:55-14:15	PVD-0409
Providence, RI	21-Apr-18	14:24-14:36	PVD-0421
Lake Road Generating Power Plant, VT	11-Apr-18	16:18-16:22	LRGPP-0411

<u>Stochastic Time-Inverted Lagrangian Transport (STILT) modeling framework and surface flux models/inventories for CO_2 source impact estimation</u>

To translate the signals observed by the aircraft to fluxes on the surface, we use observation locations that were 10 seconds apart along the ECO-2018 transects as receptors into the Stochastic Time-Inverted Lagrangian Transport Model's R interface (STILT-R version 2 [16, 17]; herewith STILT). Each STILT footprint was calculated at a 0.02° x 0.02° surface resolution, using meteorological data from the High-Resolution Rapid Refresh (HRRR) model (hourly, 3km resolution), releasing 400 particle ensembles, and simulations were extended six-hours back in time (see SI Section 1 for justification of using six hours to represent a near-field urban domain and a case-study demonstrating its validity).

STILT outputs hourly surface influence footprints ($\frac{ppm}{\mu mol/_{m^2-sec}}$) that when used in conjunction with hourly CO_2 surface fluxes ($\frac{\mu mol}{m^2-sec}$), e.g., from fossil fuel combustion or the biosphere, can estimate fossil fuel and biosphere CO_2 source impacts (ppm), respectively. Here, we convolve the footprints with one fossil fuel inventory (Anthropogenic Carbon Emissions Systems (ACES) [18]) and two biosphere models (Vegetation Photosynthesis Respiration Model (VPRM) [8, 19] and Solar-Induced Fluorescence for Modeling Urban biogenic Fluxes (SMUrF) [20]). Briefly, the ACES inventory provides fine-scale (1km x 1km), hourly fossil fuel CO_2 emissions for all major emitting sectors (e.g., power plants, on-road, commercial, residential, etc.) for 13 Northeast US states, and it has been demonstrated to be well-constrained in this region [21]. VPRM estimates ecosystem respiration and gross ecosystem exchange (GEE) at a 0.02°x0.02°, hourly resolution using a combination of Moderate Resolution Imaging Spectroradiometer (MODIS) inputs, meteorological variables (e.g., photosynthetically active radiation and temperature), and various optimized parameters. SMUrF utilizes Solar-Induced Fluorescence (SIF) as a proxy for photosynthesis and air, soil temperatures, and SIF-driven Gross Primary Production (GPP) to estimate ecosystem respiration (R_{eco}). ACES (1 km x 1 km) and SMUrF (0.05°x0.05°) were available at different resolutions than our STILT-generated footprints (0.02°x0.02°); for convolutions, we re-gridded ACES to be the same resolution and picked the nearest grid for convolutions with SMUrF.

We used two biosphere representations (VPRM and SMUrF), which use two fundamentally different approaches to represent the biosphere, to investigate the importance of its representation to the biosphere influence in studies of urban CO_2 outflows that employ a local background observing framework. The percent difference between the biosphere representations averaged within each urban core and aligned with the 6-hour back-trajectory from STILT was determined as:

$$\begin{cases} Percent\ Difference\ (\%) = \dfrac{|VPRM - SMUrF|}{(VPRM + SMUrF)/2} * 100\%, & VPRM\ and\ SMUrF\ same\ direction \\ Percent\ Difference\ (\%) = \dfrac{|VPRM - SMUrF|}{max(|VPRM|, |SMUrF|)} * 100\%, & VPRM\ and\ SMUrF\ opposing\ direction \end{cases} \qquad (Eqn. 1)$$

We use a different approach to quantify percent error if the biosphere representations are showing opposite directions (e.g., VPRM net uptake and SMUrF net respiration) to best minimize variability/error when crossing the zero flux threshold (see SI Table 1 for a comparison of percent differences between the two approaches across all urban study areas for all months). Since these two biosphere representations are drastically different, they provide an excellent proxy for evaluating how impactful the biosphere may be on urban fossil CO_2 studies within this framework – if these two representations have minimal impact on the fossil evaluation, it indicates very low sensitivity of this observing framework to the biosphere.

Quantifying daily fossil fuel and biosphere CO_2 contributions to urban outflows observed with a local background framework

For each of the model-space study domains, we perform the convolution method described above for modeled fluxes for each day of an entire calendar year. This enables us to evaluate how a flight with winds as observed during ECO-2018 that capture an urban CO_2 enhancement would sample fossil and biosphere fluxes from each urban and one point source domain over all seasons. The footprints from STILT were the same for each day to again ensure model-space would represent a real, observed urban CO_2 enhancement; we change the fossil fuel and biosphere fluxes aligned to each day. To then quantify the relative influence of the fossil and biosphere activity to a simulated urban CO_2 outflow, we used an integrated sum approach that estimates the fossil fuel and combined (i.e., fossil fuel plus biosphere) enhancement relative to simulated CO_2 levels outside of the urban core along the tails of the enhancement (Fig. 1). This approach allows us to define a local background relative of an urban enhancement, broadly consistent with the approach of defining urban outlined by Wu et al. [20, 22]. This stands in contrast to many urban study approaches using upwind tower observations, which often do not isolate the local city in the same manner, leading to the urban tower having significant sensitivity to fluxes outside of the local urban domain.

Here, the urban-defined region for each city was characterized by combining: (1) satellite imagery, (2) STILT-derived back trajectories for each model-space domain (i.e., along the simulated airborne transect, a CO_2 enhancement downwind of the urban domain could be identified against an urban-background CO_2 level on the tails, outside of the primary urban domain), and (3) local government/commission-defined urban boundaries (SI Fig. 1 for urban area designation in each of the study domains). The urban fossil fuel CO_2 enhancement is estimated as the sum of the fossil fuel enhancement relative to the simulated fossil fuel CO_2 at the tails of the enhancement (black-shaded area in Fig. 1; SI Fig. 2 for simulated CO_2 source contributions for each flight-path on the day of the flight). To account for uneven enhancements on either side of the urban-defined domain – which can be attributed to unequal fluxes due to land-use variation – we use a straight line between the tails as the local background concentration (the dashed, angled lines in Fig. 1). This value relative to the sum of the combined (fossil fuel + biosphere) enhancement (red-shaded area in Fig. 1) within the urban-defined domain can then be used to quantify the relative biosphere and fossil fuel influence to an urban outflow when observed via airborne sampling as:

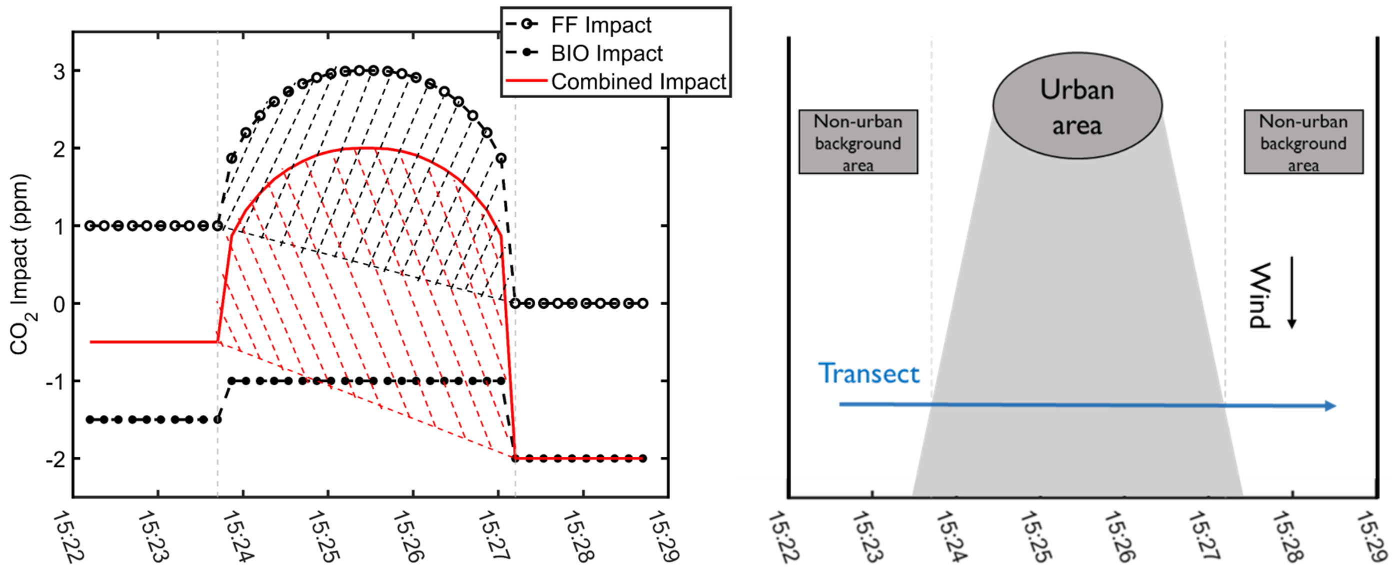
$$Biosphere\ contribution\ (\%) = 1 - \frac{urban\ fossil\ fuel\ CO_2 contribution}{combined\ (fossil\ fuel + biosphere)\ urban\ CO_2 contribution} \qquad (Eqn. 2)$$

where the fossil fuel contribution can then be determined as:

$$Fossil\ Fuel\ contribution\ (\%) = 1 - Biosphere\ contribution\ (\%) \qquad (Eqn. 3)$$

These straightforward calculations enable us to discern the relative impact of the biosphere and fossil emissions for each simulated airborne sample. These percentages directly translate into the relative importance in any follow-on analysis focused on quantifying fossil emissions. If the biosphere contributes to 10% of the total observed signal (urban-integrated combined sum), then ignoring the biosphere would only contributed to a 10% error in a fossil analysis. In contrast, if the biosphere represented 50% of the signal, neglecting it would directly translate to a 50% error.

Fig. 1: Example urban CO_2 enhancement along a downwind, airborne transect showing simplified, sample fossil fuel and biosphere contributions. (Left) The bold, red line is the combined (fossil fuel + biosphere) CO_2 enhancement. Vertical grey lines indicate the location of the urban-defined region. The shaded black area is the urban-integrated fossil fuel sum while the shaded red area is the urban-integrated combined sum. (Right) A sample visual of an airborne transect that could produce this CO_2 enhancement



Comparing airborne-observation simulated and idealized-located tower biosphere CO_2 contributions

Using the NYC-0413 transect (the only transect assessed here whose STILT-derived, 72-hour, average back-trajectory stayed in the Continental U.S.; SI Fig. 3), we treat the receptor with the highest simulated fossil fuel contribution as the idealized location of a CO_2 observing tower downwind of the urban core. This is considered the idealized location to assess an urban fossil fuel CO_2 impact as this location captures the maximum CO_2 enhancement (and presumably fossil emission signature) from the urban core (of receptors along the transect). We follow the same source-impact estimation approach using STILT described above, but instead run STILT at 0.2°x0.2° resolution, 72-hours back in time, and use VULCAN Version 3.0 fossil fuel emission inventory [23] outside of the near-field NYC domain (the ACES inventory does not include emission estimates outside of the NE US; see SI Fig. 3 for spatial domains). In the near-field NYC domain, we utilize the same CO_2 source impact results when STILT was run at a 0.02°x0.02° resolution and used ACES as described above. Conserving source contributions in the near-field domain allows us to make a more direct comparison between an idealized-located tower-observed CO_2 source contribution and one generated from an observed outflow with a local background (i.e., via a downwind airborne transect). The fossil fuel and biosphere CO_2 impacts as assessed with the idealized-located tower were compared against a 6-hour (the same time duration as the back trajectory model runs) CO_2 source impact at the same receptor and with the source contributions integrated along the entire urban outflow.

Comparing fossil fuel and biosphere CO_2 contributions from the local background observing framework approach with surface fluxes within each city

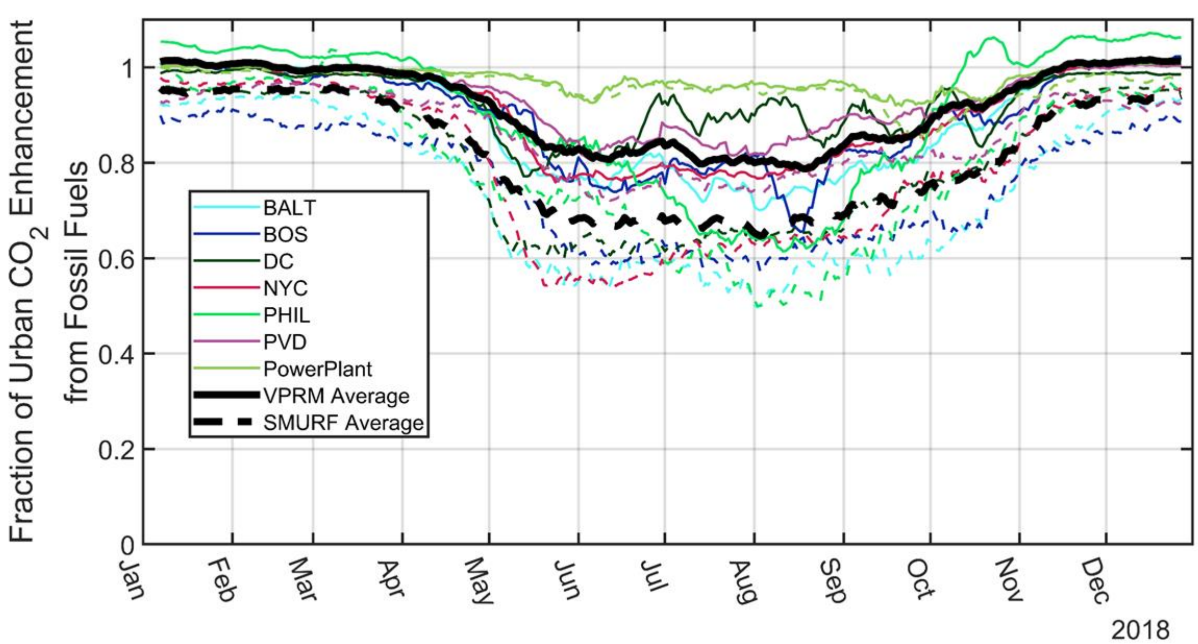
We assess surface fluxes for fossil fuel inventories (ACES and VULCAN) and biosphere models (VPRM and SMUrF) for an entire year, 2018, within each of the six urban cores in this study. For each city, we determine the sum of the flux fields within each city's urban core (SI Fig. 1) and calculate daily and 6-hour summations aligned with the same hour of day as the airborne observations (which generally represented the maximum biosphere drawdown given that most flights occurred between noon and 4:00 pm local time; Table 1). We then compare a "bottom-up" emission inventory fraction of urban CO_2 from fossil fuels following the approach outlined in Eqns. 2 and 3 (only using ACES and the 6-hour period aligned with the back trajectories of the various airborne transect times) with the findings from the local background observing framework approach (defined as "top-down") within each urban domain.

RESULTS

Annual simulated fossil fuel and biosphere CO_2 source contributions to urban outflows observed with a local background approach

Across the seven study sites (6 cities and 1 point source) and 15 unique flight paths assessed here, we find the fossil fuel CO_2 contribution always exceeds the biosphere contribution – independent of biosphere model used – when the observing technique employs a local background observing approach, i.e., via a downwind airborne transect. Outside of the summer months when a photosynthesis signal is present, the biosphere contribution is generally negligible in the modeling study presented here (Fig. 2), suggesting observing frameworks that use this local background approach under these conditions can generally ignore the biosphere and the role it plays to urban CO_2 profiles. During the summer months, the average biosphere contribution was more pronounced, with the largest monthly contribution across cities occurring in August when the average modeled biosphere contribution was 26% (range across sites: 15-36) using VPRM and 39% (23-47) using SMUrF, respectively (Fig. 2 & SI Table 2). SMUrF showed consistently higher biosphere interference compared to VPRM throughout the year, consistent with it having a stronger urban signal in each of the study cities (Table 2). The local background observing framework approach assessed here directly compares influences within an urban core to its surrounding suburban/rural-background, and SMUrF showed a larger gradient between the urban core and surrounding background throughout the year across cities (SI Fig. 4). With this observing framework, it is the magnitude of the urban-suburban-rural gradient in fluxes that drives the measured

Fig. 2: Simulated fraction of urban CO_2 from fossil fuels for each study domain (6 cities and 1 powerplant) over the course of a year using a local background observing framework (i.e., downwind airborne transect). All data is plotted as a rolling 14-day average. The solid lines represent simulations with VPRM as the biosphere representation while the dotted lines represent simulations with SMUrF. City-wide data shown here in the average of all transects in each city; see SI Fig. 5 for transect specific results within each of the study cities.



signal; even if there is a reasonably strong biosphere flux, it only impacts this observing strategy if there is a gradient in that flux from the urban core to the suburban-rural zone. The presence of a very large gradient in fossil emissions over this spatial domain creates the large urban signal, and the absence of a gradient of similar magnitude in the biosphere minimizes its impact. The 7-day rolling-average of all sites' fossil fuel contribution was slightly above one at the beginning of January and the end of December, which can be explained by a stronger relative respiration enhancement in urban-background CO_2 relative to urban levels, enhancing the observed urban CO_2 signal.

Large variability existed between VPRM and SMUrF within each of the six urban cores studied here; however, when assessing CO_2 source contributions using a local background observing framework, this variability/error was minimized (Table 2 and SI Fig. 4). The highest, monthly difference between average urban, 6-hour fluxes (aligned with the back-trajectory modeling time period) between VPRM and SMUrF averaged across the city study domains was 136% (102-180) in September where VPRM predicted an average biosphere flux of 668 (-517-1,464) nmol m^{-2} s^{-1} across cities while SMUrF modeled a flux of -524 (-1,837-106) nmol m^{-2} s^{-1} averaged across cities, where the reported fluxes are aligned with the same 6-hour interval aligned with the back-trajectory modeling. The percent difference between SMUrF and VPRM to the observed urban CO_2 enhancement when using the local background approach, on the other hand, was 15% (9.2-19). Across the summer month (June-Aug.), when the average biosphere contribution to urban CO_2 profiles is highest, and the average difference to the observed urban CO_2 enhancement when using the local background approach between the two biosphere representations was 16% (34% SMUrF and 18% VPRM averaged across cities), the percent error between biosphere models was 60% (32-116).

Table 2: Monthly comparison of biosphere fluxes (VPRM and SMUrF) across study cities that demonstrates large errors between biosphere models are minimized when assessing urban CO_2 emissions with a local background observing framework (LBOF; i.e., downwind airborne transect). The monthly average fluxes shown here correspond to the same 6-hour interval during the day that the back trajectory-modeling LBOF captures.

		Average Urban Surface Flux (nmol m^{-2} s^{-1})		**Percent Difference Between VPRM and SMUrF Urban Surface Fluxes (%)**	**Percent Difference Between VPRM and SMUrF Contribution with LBOF (%)**
		VPRM	**SMUrF**		
JAN	BALT	772	6	197	8
	BOS	627	102	144	11
	DC	699	301	80	4
	NYC	422	380	11	4
	PHIL	709	266	91	7
	PVD	363	326	11	6
	All City Average	***599***	***230***	***89***	***7***
FEB	BALT	980	237	122	7
	BOS	831	206	121	10
	DC	1058	583	58	4
	NYC	592	526	12	3
	PHIL	861	445	64	6
	PVD	516	486	2	4
	All City Average	***806***	***414***	***63***	***6***
MAR	BALT	817	-87	111	10
	BOS	547	170	105	12
	DC	783	186	123	4
	NYC	458	344	28	4
	PHIL	751	168	127	4
	PVD	402	303	28	4
	All City Average	***626***	***181***	***87***	***6***
APR	BALT	634	-366	158	15
	BOS	546	-254	146	10
	DC	606	-9	101	10
	NYC	444	240	60	7
	PHIL	530	-83	116	15
	PVD	401	168	82	5

	All City Average	***527***	***-51***	***111***	***11***
MAY	BALT	-1195	-2897	83	20
	BOS	-1984	-2669	29	16
	DC	-1089	-1660	42	20
	NYC	-443	-865	65	21
	PHIL	-653	-1442	75	11
	PVD	-1058	-463	78	9
	All City Average	***-1070***	***-1666***	***62***	***16***
JUNE	BALT	-2148	-4710	75	21
	BOS	-4259	-5853	32	15
	DC	-1646	-2814	52	21
	NYC	-1487	-1902	25	22
	PHIL	-1874	-3059	48	11
	PVD	-2640	-1283	69	9
	All City Average	***-2342***	***-3270***	***50***	***17***
JUL	BALT	-1864	-3427	59	19
	BOS	-3354	-4268	24	19
	DC	-1349	-1957	37	26
	NYC	-995	-1667	50	16
	PHIL	-1463	-2474	51	8
	PVD	-2338	-685	109	8
	All City Average	***-1894***	***-2413***	***55***	***16***
AUG	BALT	-1082	-2360	74	18
	BOS	-1552	-2691	54	13
	DC	-1122	-1048	7	24
	NYC	-504	-726	36	15
	PHIL	-342	-1293	116	12
	PVD	-993	-82	169	8
	All City Average	***-933***	***-1367***	***76***	***15***
SEP	BALT	1219	-772	163	19
	BOS	-517	-1837	112	18
	DC	1250	-21	102	15
	NYC	723	-255	135	14
	PHIL	1464	-364	125	13
	PVD	-133	106	180	9
	All City Average	***668***	***-524***	***136***	***15***
OCT	BALT	1173	-534	146	21

	BOS	593	-812	173	20
	DC	1122	-7	101	13
	NYC	674	40	178	13
	PHIL	766	-111	115	21
	PVD	381	224	52	9
	All City Average	***785***	***-200***	***127***	***16***
NOV	BALT	1049	-31	103	15
	BOS	918	128	151	14
	DC	973	279	111	5
	NYC	683	415	49	7
	PHIL	940	207	128	12
	PVD	678	349	64	6
	All City Average	***874***	***225***	***101***	***10***
DEC	BALT	956	549	54	8
	BOS	759	495	42	13
	DC	862	759	13	3
	NYC	577	692	18	5
	PHIL	846	709	18	11
	PVD	513	561	9	8
	All City Average	***752***	***627***	***26***	***8***

Comparing fossil fuel and biosphere CO_2 source contributions between idealized-located tower and local background observing framework

CO_2 source contributions to the idealized-located tower showed a large relative biosphere contribution throughout the year (respiration signal in winter and drawdown in summer months) compared to fossil fuel contributions (Fig. 3a), consistent with the findings from previous tower-network studies of urban CO_2 that suggest the biosphere is an important parameter when assessing urban CO_2 [5, 6]. However, when utilizing a local background observing framework, the biosphere impact, although still present, is considerably less pronounced (Fig. 3b for 6-hour back from the idealized-located tower and Fig. 3c for the source contribution along the entire NYC-0413 transect). During peak biosphere CO_2 contributions (June 4-10) of the 72-hour back trajectories to the idealized-located tower along the NYC-0413 downwind flight path, we find the biosphere contribution from VPRM and SMUrF exceeds the fossil contribution, with 7-day rolling average contributions of 59.1% and 73.2% to the urban CO_2 enhancement, respectively. During this same period, using the local background observing framework approach, the biosphere represents just 9.8% and 28.6% of the simulated CO_2 profile, respectively (Fig. 3c). These findings further suggest that while the biosphere is important to characterize in certain observing frameworks, when urban outflows are measured with a local background, as facilitated by airborne or satellite-based observations, the biosphere is less important to characterize and errors in its representation will not compound in studies of urban fossil fuel CO_2.

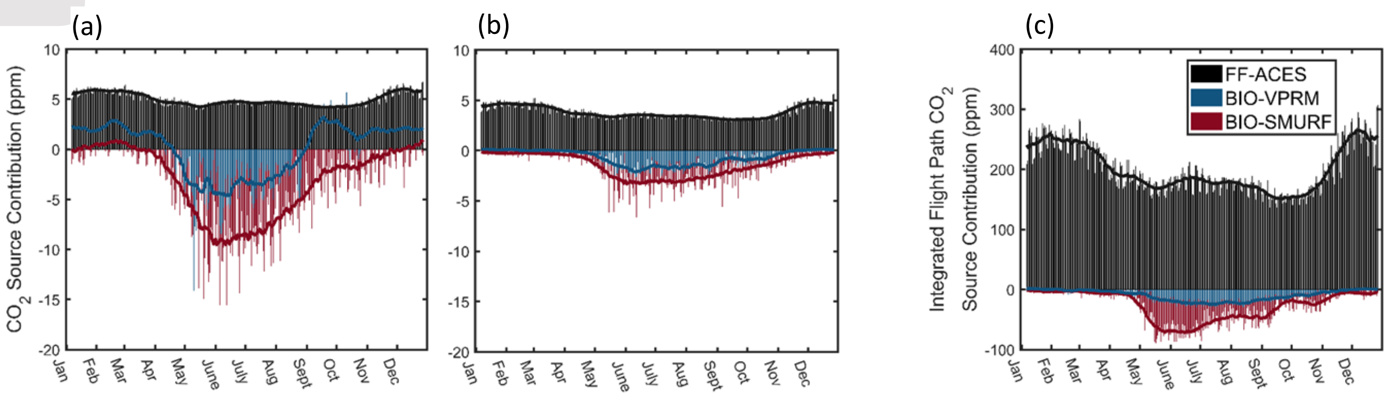
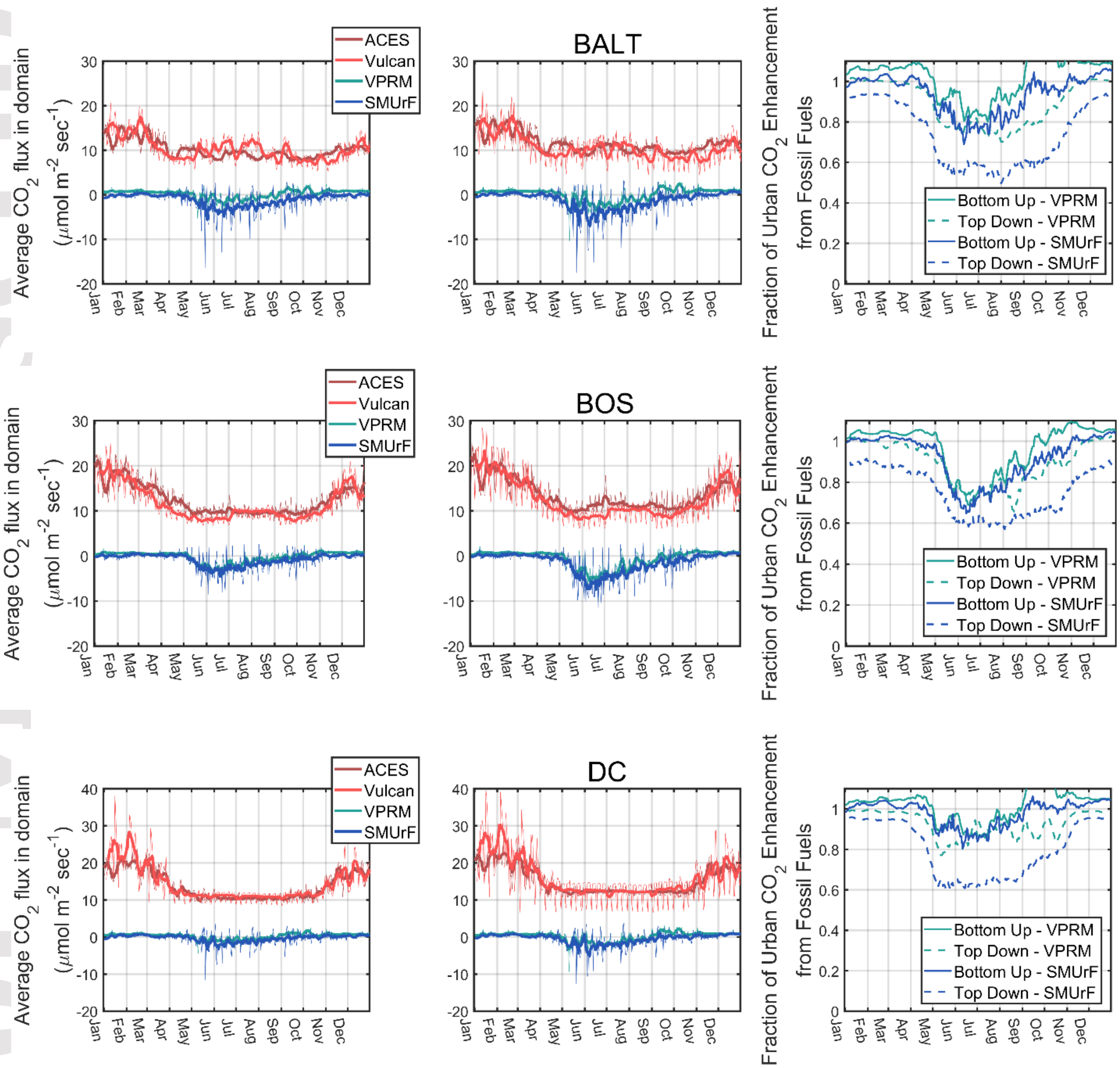


Fig. 3: Comparison of fossil fuel and biosphere CO_2 contributions to: (a) Idealized-located tower convolved 72 hours back in time; (b) The same idealized-located tower convolved six hours back in time (the same duration of the back trajectory modeling convolution); and (c) Along the entire urban outflow for the NYC-0413 transect. Seven-day rolling averages for both fossil fuel and biosphere source contributions are indicated by the bold lines.

Bottom-up fossil fuel and biosphere CO_2 surface flux comparison with top-down local background approach to assess CO_2 source contributions

For each of the six cities assessed here, fossil fuel CO_2 emissions have higher magnitudes than CO_2 signals from the biosphere throughout the year (Fig. 4, left column), even during afternoon periods (6-hour period aligned with downwind airborne observation back-trajectory modeling time period) when the biosphere drawdown would be more pronounced (Fig. 4, middle column). When comparing bottom-up fluxes against top-down local background CO_2 source contributions, we find that across cities, the bottom-up approach generally showed a lower biosphere influence than the top-down approach (Fig. 4, right column & Fig. 5) for both biosphere representations. This is attributed to the difference in what each approach is directly assessing: the emission inventory approach only assesses direct urban fluxes while the flight-path approach is geared toward assessing enhancements relative to a local background – it's capturing the biosphere gradient along the urban-suburban-rural transect. The biosphere gradient between the urban core and its surrounding (SI Fig. 4) results in the increased relative contribution of the biosphere to a simulated urban CO_2 enhancement. We further find that SMUrF generally resulted in a larger biosphere contribution compared to VPRM with both the bottom-up and top-down approaches, consistent with it having a stronger signal within each of the urban cores assessed here and a steeper urban-suburban-rural gradient, respectively (Table 2).


BALT
BOS
DC
ACES
Vulcan
VPRM
SMUrF
Average CO2 flux in domain (μmol m-2 sec-1)
Fraction of Urban CO2 Enhancement from Fossil Fuels
Bottom Up - VPRM
Top Down - VPRM
Bottom Up - SMUrF
Top Down - SMUrF
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

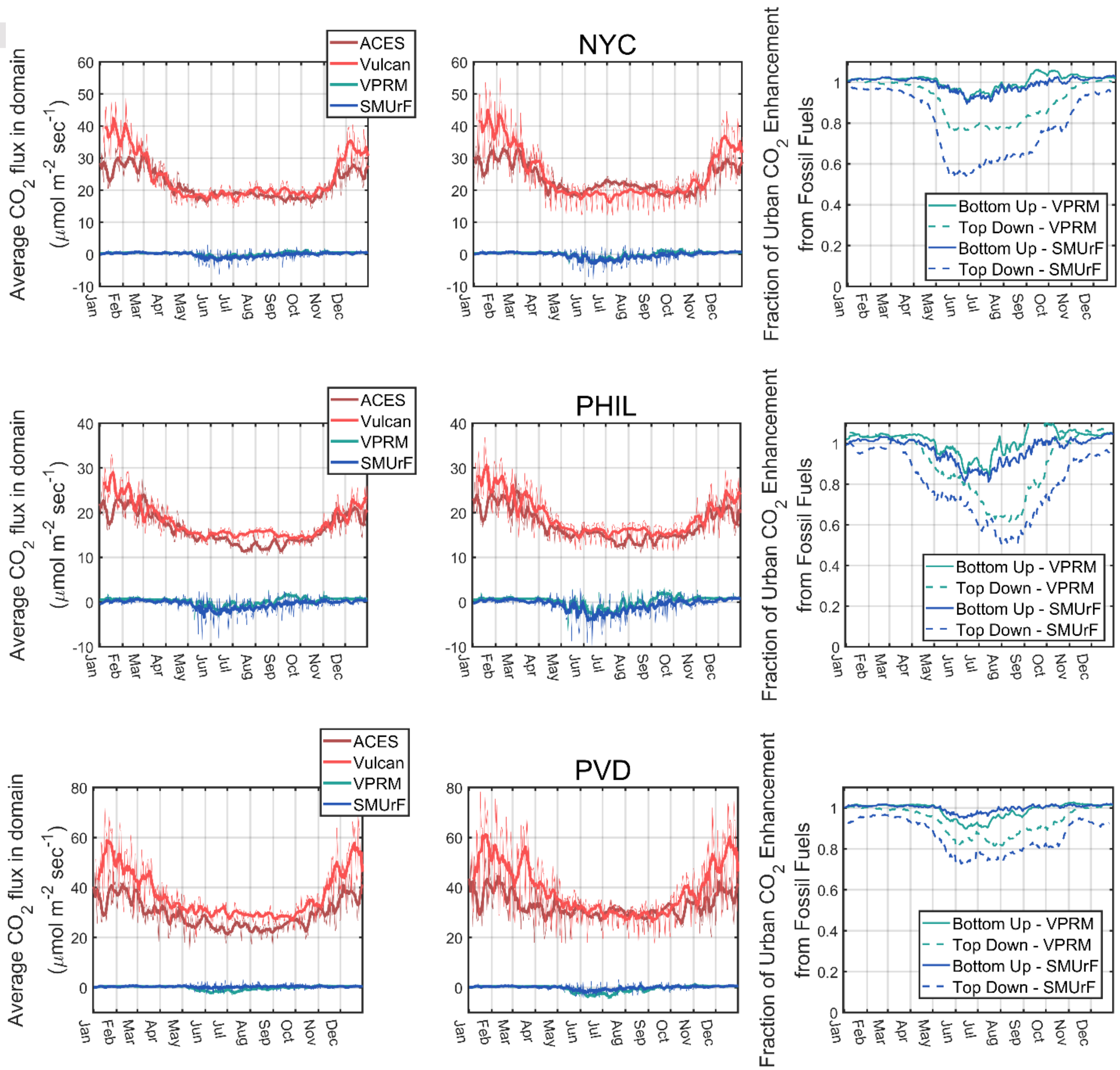


Fig. 4: Comparison of fossil fuel and biosphere surface fluxes (µmol m^{-2} s^{-1}) across the six study cities for (left column) daily-average within the domain and (middle column) 6-hour average (aligned with back-trajectory modeling time domain) within the domain. (Right column) Comparison of the fraction of urban CO_2 from fossil fuels between bottom-up, emission inventory and top-down, local background observing framework (i.e., downwind airborne transect).

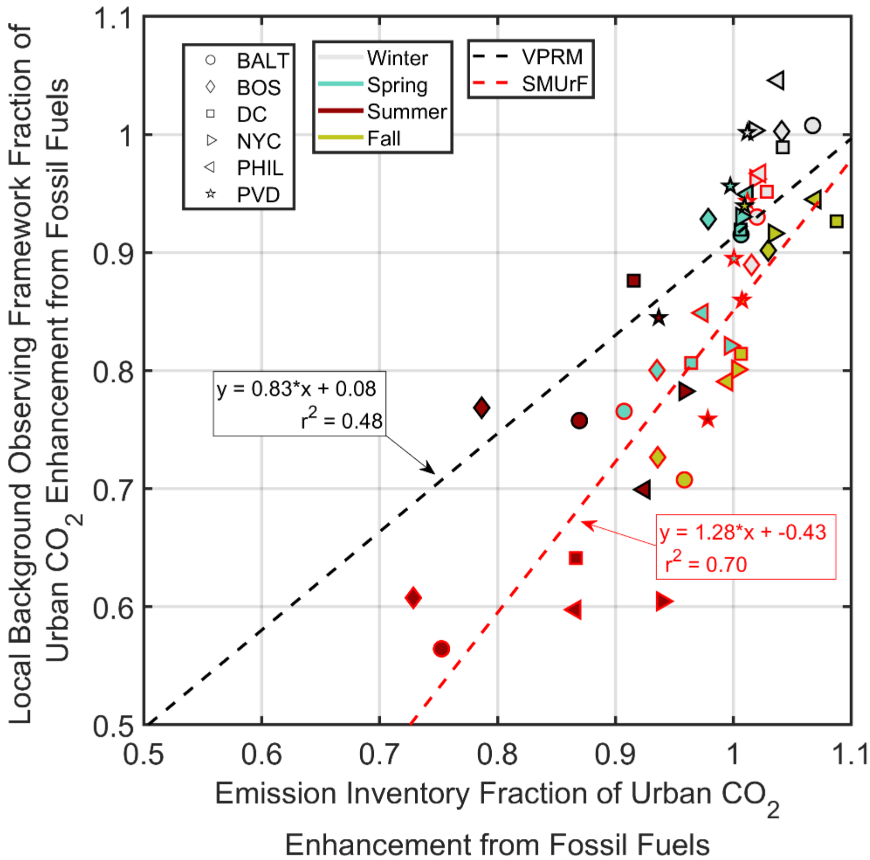


Fig. 5: Seasonal comparison of bottom-up emission inventory and top-down local background observing framework approach (i.e., downwind airborne transect) approach to quantify the fraction of urban CO_2 from fossil fuels across the six study cities and two biosphere representations (VPRM and SMUrF).

<u>DISCUSSION, LIMITATIONS, & CONCLUSIONS</u>

Cities in the United States have spearheaded nationwide efforts for deep de-carbonization efforts, including the introduction of ambitious CO_2 emission reduction pathways. Observation-based evaluations of fossil fuel CO_2 emissions can be influenced by the biosphere, which can act as both a source (respiration) and sink (photosynthesis) of CO_2; thus, understanding and characterizing the role of the biosphere to studies of urban CO_2 can help constrain fossil emissions in cities. A frequently used approach to characterize urban CO_2 is to analyze tower (or tower-network) observations, and although this approach can provide a long temporal record, the spatial sensitivity of these stationary sensing platforms are dictated by meteorological conditions and inversion approaches that often capture air masses that travel distances well upwind of a direct urban core, potentially inflating the role the biosphere plays to urban CO_2 profiles. For example, a previous tower-based study in Boston, MA. concluded that biosphere activity offset 100%, 58%, and 20% of the afternoon anthropogenic urban CO_2 enhancement in July, September, and October, respectively [6]. In addition, using a network of towers in Indianapolis, IN. as part of the INFLUX campaign, Turnbull et al. [13] showed the biosphere contributes >50% of urban CO_2 signals in summer months, while Miller et al. [5], also using multiple towers in Los Angeles, CA., concluded that the urban biosphere is 33% of the annual mean fossil fuel contribution – a surprisingly large contribution for a dry, metropolitan area.

In this study, we estimate the influence of the biosphere to cities' CO_2 profiles using a back-trajectory modeling approach that employs a local background observing framework, consistent with what would occur if a city's CO_2 outflow was measured via airborne or satellite-based observations. We find that studies of urban CO_2 that employ such a local background approach substantially minimizes potential biosphere interference to urban CO_2 profiles – across study sites here, the biosphere contribution to the total CO_2 signal was 15% across the year (<10% in non-summer months), so in studies conducted outside of summer months that use this observing framework, it can be concluded that errors in fossil fuel emission estimates will be <10% due to biosphere interference, suggesting this approach can be used to conduct robust assessments of urban fossil fuel CO_2 emissions. Similar space-based observation strategies that capture local background will have similar success. For ground-based observations, capturing the local background will have similar success. For ground-based observations, capturing the local background across varied wind conditions becomes more challenging, and can greatly increase the number of observations needed to minimize the biosphere influence. Dense networks of surface-level, lower-cost monitors [24] could potentially be leveraged to assess urban fossil fuel emissions with observations of local-background to attempt to minimize biosphere influences. This does not imply sparse urban CO_2 observing networks do not provide additive information on CO_2 fossil emissions, but merely that this observing strategy has greater sensitivity to biosphere fluxes.

In addition, the local background observing framework minimizes errors in how well the biosphere needs to be represented in studies of urban CO_2. Here, we used two biosphere models that had an average bias of >80%; but the difference of impact a simulated CO_2 enhancement, on average, was ~12%, further demonstrating the efficacy of this observing approach to characterizing urban fossil fuel emissions. Outside of summer months, the biosphere can generally be ignored in studies of urban CO_2 that utilize such a local background observing framework, providing opportunities to perform robust studies of fossil fuel emissions within cities.

Model simulations for each study domain were assessed in the afternoon, consistent with the timing of most airborne campaigns, which are scheduled for times with developed boundary layer heights to best capture urban emissions and be less subject to long-range transport. In addition, this approach allows for comparison to satellite retrievals whose overpasses occur in the afternoon and which can be used to sample the urban domain considering a local background much like the flight-based sampling approach. Further, the time of the airborne transects as assessed here generally aligns with the period when the biosphere drawdown will be more pronounced, suggesting the findings presented here capture a high biosphere contribution to urban CO_2 from the local background approach. An additional advantage of using this local background approach to isolating fossil fuel emissions is the observations do not have to be directly along the edge of a city, but can have additional biosphere interferences between the observing points and urban core as this approach is driven by defining the urban-suburban-rural gradient, unlike in single-tower based studies where that additional biosphere contribution would further promote the amplified role of the biosphere to urban CO_2.

Cities around the world, which are often associated with large carbon footprints, have announced targets to reduce emissions and transition to net-zero economies across their supply chains. Using top-down atmospheric observations has become a critical tool to evaluate such emissions; however, for CO_2 specifically, these inversion approaches are subject to interferences from the biosphere, which can act as both a source and sink of CO_2. Here, using high-resolution, back-trajectory modeling approaches, we show that using observing frameworks that incorporate a local background (e.g., via airborne or space-based observations) can minimize biosphere interference to urban CO_2 for estimate whole city emissions, paving the way to conduct robust studies of urban fossil fuel CO_2 emissions to ultimately reduce emissions and combat climate change.

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OPEN RESEARCH

Data Availability Statement:

Aircraft observation data as part of the ECO flight campaign is available at **ftp://aftp.cmdl.noaa.gov/data/campaign/ECO_2018/ECO_2018_insitu_10sec.nc.** STILT back-trajectory model source code is available for downloaded at **https://uataq.github.io/stilt/#/**. ACES emission inventory is available from the ORNL DAAC at **https://daac.ornl.gov/CMS/guides/CMS_Carbon_Emissions_NE_US.html**. VPRM hourly flux data is available at **https://data.nist.gov/od/id/mds2-2382**. SMUrF hourly flux data is available from the ORNL DAAC at **https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1899**. HRRR met data is available at **https://rapidrefresh.noaa.gov/hrrr/**

WORKS CITED

1. Ramaswami, A., et al., *Carbon analytics for net-zero emissions sustainable cities.* Nature Sustainability, 2021. **4**(6): p. 460-463.
2. Mi, Z., et al., *Cities: The core of climate change mitigation.* Journal of Cleaner Production, 2019. **207**: p. 582-589.

3. Dodman, D., *Blaming cities for climate change? An analysis of urban greenhouse gas emissions inventories.* Environment and urbanization, 2009. **21**(1): p. 185-201.
4. Gurney, K.R., et al., *Under-reporting of greenhouse gas emissions in U.S. cities.* Nature Communications, 2021. **12**(1): p. 553.
5. Miller, J.B., et al., *Large and seasonally varying biospheric CO₂ fluxes in the Los Angeles megacity revealed by atmospheric radiocarbon.* Proceedings of the National Academy of Sciences, 2020. **117**(43): p. 26681-26687.
6. Sargent, M., et al., *Anthropogenic and biogenic CO₂ fluxes in the Boston urban region.* Proceedings of the National Academy of Sciences, 2018. **115**(29): p. 7491-7496.
7. Hardiman, B.S., et al., *Accounting for urban biogenic fluxes in regional carbon budgets.* Science of The Total Environment, 2017. **592**: p. 366-372.
8. Gourdji, S.M., et al., *A Modified Vegetation Photosynthesis and Respiration Model (VPRM) for the Eastern USA and Canada, Evaluated With Comparison to Atmospheric Observations and Other Biospheric Models.* Journal of Geophysical Research: Biogeosciences, 2022. **127**(1): p. e2021JG006290.
9. Karion, A., et al., *Background conditions for an urban greenhouse gas network in the Washington, DC, and Baltimore metropolitan region.* Atmos. Chem. Phys., 2021. **21**(8): p. 6257-6273.
10. Kort, E.A., et al., *Surface observations for monitoring urban fossil fuel CO2 emissions: Minimum site location requirements for the Los Angeles megacity.* Journal of Geophysical Research: Atmospheres, 2013. **118**(3): p. 1577-1584.
11. Yadav, V., et al., *The Impact of COVID-19 on CO2 Emissions in the Los Angeles and Washington DC/Baltimore Metropolitan Areas.* Geophysical Research Letters, 2021. **48**(11): p. e2021GL092744.
12. Karion, A., et al., *Greenhouse gas observations from the Northeast Corridor tower network.* Earth Syst. Sci. Data, 2020. **12**(1): p. 699-717.
13. Turnbull, J.C., et al., *Synthesis of Urban CO2 Emission Estimates from Multiple Methods from the Indianapolis Flux Project (INFLUX).* Environmental Science & Technology, 2019. **53**(1): p. 287-295.
14. McKain, K., et al., *Assessment of ground-based atmospheric observations for verification of greenhouse gas emissions from an urban region.* Proceedings of the National Academy of Sciences, 2012. **109**(22): p. 8423-8428.
15. Plant, G., et al., *Large Fugitive Methane Emissions From Urban Centers Along the U.S. East Coast.* Geophysical Research Letters, 2019. **46**(14): p. 8500-8507.
16. Fasoli, B., et al., *Simulating atmospheric tracer concentrations for spatially distributed receptors: updates to the Stochastic Time-Inverted Lagrangian Transport model's R interface (STILT-R version 2).* Geosci. Model Dev., 2018. **11**(7): p. 2813-2824.
17. Lin, J.C., et al., *A near-field tool for simulating the upstream influence of atmospheric observations: The Stochastic Time-Inverted Lagrangian Transport (STILT) model.* Journal of Geophysical Research: Atmospheres, 2003. **108**(D16).
18. Gately, C. and L. Hutyra, *CMS: CO2 emissions from fossil fuels combustion, ACES Inventory for Northeastern USA.* ORNL DAAC, 2018. **10**.
19. Mahadevan, P., et al., *A satellite-based biosphere parameterization for net ecosystem CO2 exchange: Vegetation Photosynthesis and Respiration Model (VPRM).* Global Biogeochemical Cycles, 2008. **22**(2).
20. Wu, D., et al., *A model for urban biogenic CO2 fluxes: Solar-Induced Fluorescence for Modeling Urban biogenic Fluxes (SMUrF v1).* Geosci. Model Dev., 2021. **14**(6): p. 3633-3661.

21. Gately, C.K. and L.R. Hutyra, *Large Uncertainties in Urban-Scale Carbon Emissions.* Journal of Geophysical Research: Atmospheres, 2017. **122**(20): p. 11,242-11,260.
22. Wu, D., et al., *A Lagrangian approach towards extracting signals of urban CO2 emissions from satellite observations of atmospheric column CO2 (XCO2): X-Stochastic Time-Inverted Lagrangian Transport model ("X-STILT v1").* Geosci. Model Dev., 2018. **11**(12): p. 4843-4871.
23. Gurney, K.R., et al., *The Vulcan Version 3.0 High-Resolution Fossil Fuel CO2 Emissions for the United States.* Journal of Geophysical Research: Atmospheres, 2020. **125**(19): p. e2020JD032974.
24. Shusterman, A.A., et al., *The BErkeley Atmospheric CO2 Observation Network: initial evaluation.* Atmos. Chem. Phys., 2016. **16**(21): p. 13449-13463.