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Policy-Relevant Assessment of Urban CO_2 Emissions

Thomas Lauvaux,* Kevin R. Gurney, Natasha L. Miles, Kenneth J. Davis, Scott J. Richardson, Aijun Deng, Brian J. Nathan, Tomohiro Oda, Jonathan A. Wang, Lucy Hutyra, and Jocelyn Turnbull

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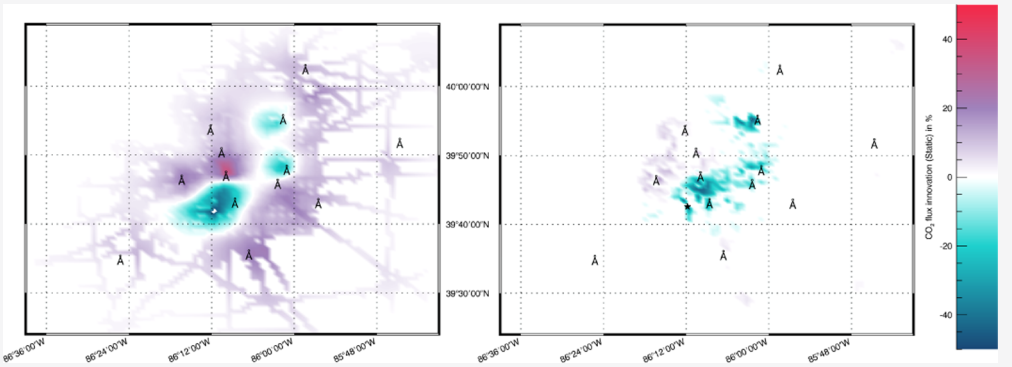

ABSTRACT: Global fossil fuel carbon dioxide ($FFCO_2$) emissions will be dictated to a great degree by the trajectory of emissions from urban areas. Conventional methods to quantify urban $FFCO_2$ emissions typically rely on self-reported economic/energy activity data transformed into emissions via standard emission factors. However, uncertainties in these traditional methods pose a roadblock to implementation of effective mitigation strategies, independently monitor long-term trends, and assess policy outcomes. Here, we demonstrate the applicability of the integration of a dense network of greenhouse gas sensors with a science-driven building and street-scale $FFCO_2$ emissions estimation through the atmospheric CO_2 inversion process. Whole-city $FFCO_2$ emissions agree within 3% annually. Current self-reported inventory emissions for the city of Indianapolis are 35% lower than our optimal estimate, with significant differences across activity sectors. Differences remain, however, regarding the spatial distribution of sectoral $FFCO_2$ emissions, underconstrained despite the inclusion of coemitted species information.

INTRODUCTION

Despite the recent increase in renewable energy supply and the transition from coal to natural gas, fossil fuel emissions of carbon dioxide ($FFCO_2$) continue to increase globally at a rate of about 2% annually, a pace driven primarily by growth in Asia and the United States (+2−3%) somewhat moderated by Europe (−2.5%) in 2018.[1] Roughly three-quarters of energy-related $FFCO_2$ emissions are generated by urban-based activities[2] implying that subnational and city governments have the opportunity to play a central role in achieving the objectives of emission reductions as stated by the Paris agreement[a].[3] Trends in global population distribution suggest that urban growth will be dominated by large and medium-sized urban areas in the coming decades (≥250 000 people), projected to represent 70% of the global population by 2050 compared to 50% in 2016.[4] In response to the challenges of the growing energy demand and in support of international climate agreements, city governments and consortia of major metropolitan areas have begun to pledge and implement mitigation strategies (e.g., C40 Cities Climate Leadership Group[b]). $FFCO_2$ emission reduction choices, cost-effectiveness, and reduction assessment depend on having a reliable quantitative understanding of urban emissions with sufficient spatial and functional detail and a means to assess trends[5] and individual reduction policy effectiveness.[6] Despite efforts by city governments to build self-reported inventories, emission uncertainties remain large and likely exceed annual urban emission trends in emission reductions (which leveled at about 2% per year for the top third of the C40 cities).[7] Emission quantification at the subcity scale and useful integration with cobenefits such as air quality goals in addition to generating targeted and cost-effective reduction options[8] remain particularly challenging for cities to generate in budget-constrained conditions. Finally, standardized data sources and estimation procedures are not fully developed and/or adopted leading to continued discrepancies among inventory estimates.[9] The role of atmospheric measurements to monitor long-term trends in

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greenhouse gas (GHG) fluxes has been unequivocal at the global scale.[10] A similar approach to quantifying fluxes and flux trends has been suggested at finer scales, with a particular emphasis on cities, given the increasing policy demand and the autonomy many cities possess over GHG reduction policy suggests and opportunity for new mitigation action.[11] Approaches that combine or assimilate information from a collection of observed quantities to constrain fluxes at relevant scales in urban domains are in rapid development.[12,13] Atmospheric approaches assimilate atmospheric mixing ratio measurements and advanced bottom-up flux estimation into models of urban atmospheric transport to achieve the required granularity in emissions and hence become policy relevant.[14−17] However, most studies have focused on annual whole-city emissions lacking information on individual sectors of the economy. The combination of integrated flux accuracy with space/time and process (e.g., fuel, sector, combustion type, vegetation type) flux detail offers relevant and reliable information for urban stakeholders.[18] Such an approach might achieve the level of confidence required to assist local governments to make informed decisions and track their progress toward mitigation goals. To achieve levels of precision sufficient to assess sector-specific emission trends from various sectors of the economy and do so with spatial detail, the approach requires a dense network of atmospheric sensors and space/time-explicit bottom-up emissions estimates. Among the many ongoing experiments testing these approaches in urban areas, an experiment deployed in 2010, the Indianapolis Flux Experiment (INFLUX),[19] provides an ideal test-bed for thoroughly exploring specific aspects of this approach, namely the density and accuracy of the atmospheric carbon (CO_2, CH_4, CO) monitoring component,[20] the contribution of high-resolution bottom-up $FFCO_2$ emission estimates,[21] urban vegetation flux estimation,[22] and the sensitivity of the inversion setup.[16]

With 872 680 inhabitants, Indianapolis ranks 17th among US cities[c] and 287th globally. Among city populations in developed and developing countries, Indianapolis stands in the top third of global metropolitan areas, composed of 500 000−5 million inhabitants. About 20% of these urban areas are larger than 5 million (about 70 metropolitan areas around the world), while 45% of the world population lives in cities of 300 000 or less. Hence, medium-sized cities like Indianapolis represent a baseline for monitoring systems, assuming a significant fraction of the global fossil fuel emissions would be monitored by atmospheric systems. With productive agricultural land, limited forests, and an expansive urbanized area, Indianapolis is a fair representation of metropolitan areas located in the mid-latitudes and an important first step in demonstrating the potential of advanced assimilation approaches to support climate change decision-making in cities.

Consideration of the physical, economic, and demographic conditions of Indianapolis and its relationship to the broader landscape is crucial to building the experiment and interpreting the results. Metropolitan areas in the Midwest of the United States are typically organized around a medium-density downtown, encircled by a traffic-heavy beltway itself connected to major interstate highways with large amounts of commercial truck traffic. Such is the case of Indianapolis, IN with emissions from on-road vehicles adding up to half of the city budget.[21] Suburban areas are spread unevenly in development blocks, forming concentric layers of decreasing housing density, correlated with the distance from the beltway. Industrial facilities are distributed somewhat randomly within and around the central urban area with the notable presence of a major power plant (Harding Street) that releases a third of the city emissions. Mixed vegetation (e.g., vegetation type, management level) exists throughout the city with increasing density in the outer ring of primarily suburban development.

The advanced assimilation approach described here remains nontrivial to implement at the city scale. Multiple challenges have to be addressed such as the collection of atmospheric carbon measurements at selected locations and at a sufficient height in and around the city,[20] the representation of the local atmospheric dynamics,[23] construction of the optimization algorithm used to assess $FFCO_2$ emissions separately from biogenic carbon exchange, and construction of the building/street-scale bottom-up $FFCO_2$ emissions data product.[21] Here, we demonstrate how advanced assimilation approaches can detect subcity fluxes at sufficiently small uncertainty to track potential mitigation policies by associating atmospheric information with emissions from various data sources on human activities. Our findings are based on a state-of-the-art optimization system combining multiple atmospheric trace gases and two broad sectors of economic activity.

METHODS

The optimization system presented here assimilates hourly atmospheric mixing ratios of CO_2 and CO collected during the afternoon (12 p.m. to 5 p.m.) with anthropogenic and biogenic surface fluxes at 1 km resolution. Tower footprints representing the relationship between mixing ratios and surface fluxes were simulated using simulations from the Weather Research and Forecasting model (WRFv3.6.1[24]) in Four-Dimensional Data Assimilation mode,[23] assimilating surface stations of the World Meteorological Organization, coupled offline to the Lagrangian Particle Dispersion Model[25] in backward mode. Particles were released at 20 s intervals (6300 per hour per tower) and stored every 2 min to track air motion at 1 km resolution. The optimization uses a Bayesian inversion framework[16] solving analytically for a 5 day mean of three flux components: stationary and mobile anthropogenic emissions, and biogenic fluxes. Attribution of flux contributions to CO is based on emission factors developed by adjusting reported sectoral emission factors to match flask-based observations of $^{14}CO_2$, CO_2, and CO from the INFLUX tower network.[26,27] The first guess (or state vector) represents the three flux components at 1 km resolution from the Hestia emission product[21] for emissions and from a modified version of VPRM[17] for biogenic fluxes.

Atmospheric mixing ratio measurements were collected at 12 tower locations for CO_2 and a subset of five sites for CO mixing ratios, averaged hourly from calibrated Picarro Cavity Ring-Down Spectrometers.[20,28,29] Air samples were dried to below 0.2% H_2O (0.6% at Site 02) to avoid the need for yearly water vapor calibrations.[30] Calibration protocol to comply with WMO/NOAA standards[31] was evaluated to reach an accuracy of 0.18 ppm for CO_2 and 6 ppb for CO when compared to discrete flask samples.[29] Observations were made at heights ranging from 39 to 136 m above ground level using existing communication towers.[20] Data availability and site locations are described in SI Appendix.

Bottom-up emissions from the Hestia CO_2 emission product[21,32] are available for each of the eight economic sectors, merged into two main sectors of emissions[27] corresponding to low (referred to as stationary incl. residential,

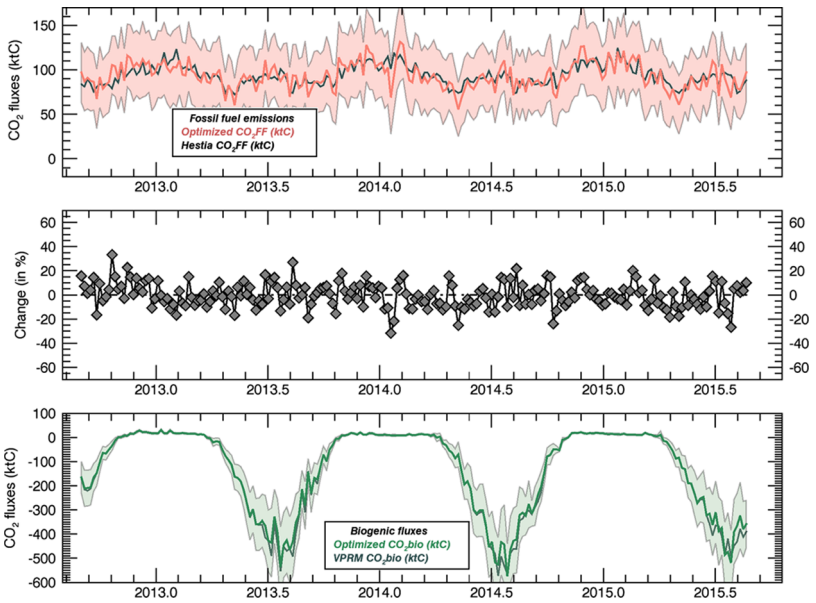

Figure 1. Whole-city 5 day averaged $FFCO_2$ emissions from the city of Indianapolis, IN (nine counties) in ktC from September 2012 to September 2015 before and after optimization for the fossil fuel CO_2 emissions (upper panel), the biogenic fluxes (lower panel), and the adjustments after optimization (difference between Hestia and the optimized emissions) (middle panel), all aggregated from 1 km resolution fluxes.

industrial, commercial, utility, airport, and railroad sectors) and high (ref to as mobile incl. on-road and off-road sectors) CO/CO_2 ratios.[33] Hestia emissions were aggregated from the initial building-level product down to the 1 km resolution footprint grid, covering Marion County and the eight surrounding counties.

The Open-source Data Inventory for Anthropogenic CO_2 (ODIAC) spatial distributions were estimated at 1×1 km^2 resolution.[34] Compared to Hestia, ODIAC is available globally by downscaling national fossil fuel consumption statistics (cf. Supporting Information), hence applicable to other urban areas across the world. The emissions from power plants are mapped using the geolocation reported in the Carbon Monitoring and Action (CARMA) global power plant database,[d] and the rest of the emissions (nonpoint source emissions) are distributed using satellite observed nightlight data. When using ODIAC as a priori, only the total fossil fuel emissions (no sector) and biogenic fluxes were optimized. A single CO/CO_2 ratio was used corresponding to the weighted average over the eight sectors of Hestia.

The urban Vegetation Photosynthesis and Respiration Model[35] incorporates the highly variable fraction of impervious surface area (ISA) from the National Land Cover Database (NLCD[36]) to modulate carbon fluxes from these heterogeneous landscapes. Nonpaved portions of Indianapolis were defined as deciduous, broadleaf forest. VPRM is driven by greenness data from the MODIS product and NARR climate reanalysis, while parameters were optimized for four land cover/land use types (i.e., irrigated maize, soybeans, grasslands, and forest)[37] to produce hourly carbon fluxes as the weighted average of carbon fluxes for each of the four land cover/land use classes.

We defined uncertainties in the Hestia and ODIAC fossil fuel emissions with variances proportional to the net emissions (100% of the net pixel emissions) and spatial error correlations (i.e., error covariances) following urban land cover and an exponentially decaying length scale of 4 km.[16] Sensitivity experiments showed that only uncertainties in Harding Street power plant emissions had a significant impact on the spatial attribution (cf. Supporting Information). For the VPRM biogenic fluxes, the uncertainties were assumed to be larger than fossil fuel emissions as diagnosed from previous studies.[37] An exponentially decaying length scale of 12 km was assumed over 5 days over the vegetated area. Transport model uncertainties were defined at each hour based on model-data statistics of wind speed and direction,[23] further calibrated based on a χ^2 normalized distance between 0.75 and 1.25.[16] Final uncertainties on fossil fuel emissions and biogenic fluxes presented in this study correspond to the posterior error covariances produced by the inversion system aggregated over the domain.

RESULTS

Building- and street-level emissions from the Hestia Project[21,32] serve as an adjustable bottom-up flux constraint to the optimization system, which sequentially assimilates daytime hourly CO_2 and CO atmospheric mixing ratios from the INFLUX network over 5 day periods. The optimization procedure generates 1 km resolution maps of sectoral CO_2 emissions for mobile and stationary sources and biogenic fluxes from vegetation. Over 3 years (September 2012−September 2015), aggregated whole-city emissions are only slightly lower than the Hestia estimate (−0.6%) with minor adjustments to 5 day $FFCO_2$ emissions (14%; cf. Figure 1). Biosphere net carbon uptake, based on simulated ecosystem exchanges from the Vegetation Photosynthesis and Respiration Model (VPRM;[17,35,38] SI Appendix, Section 5) modified to simulate urban and agricultural land covers, decreased by 7% over the 3 years, with winters showing a 3−8% reduction in net emission (less positive flux) and summers showing a 6−8% reduction in net uptake (less negative). With a larger biogenic flux adjustment in summer (reduced photosynthetic uptake), the 3 year adjustment corresponds to an overall decrease of the biogenic net carbon uptake. $FFCO_2$ emissions were little adjusted in summer (<0.5%) due to the tighter uncertainty constraint on $FFCO_2$ emissions compared to the biogenic

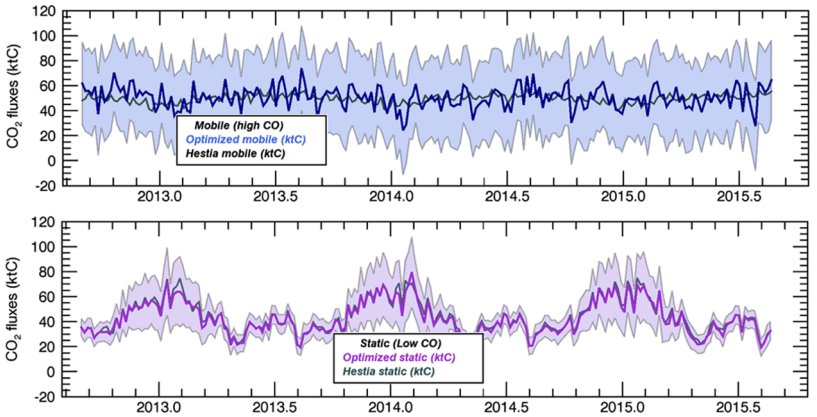

Figure 2. Whole-city 5 day averaged $FFCO_2$ emissions from the city of Indianapolis, IN (nine counties) in ktC from September 2012 to September 2015 before and after optimization for each sector of the fossil fuel emissions with mobile (upper panel) and stationary (lower panel) sources, all aggregated from 1 km resolution fluxes.

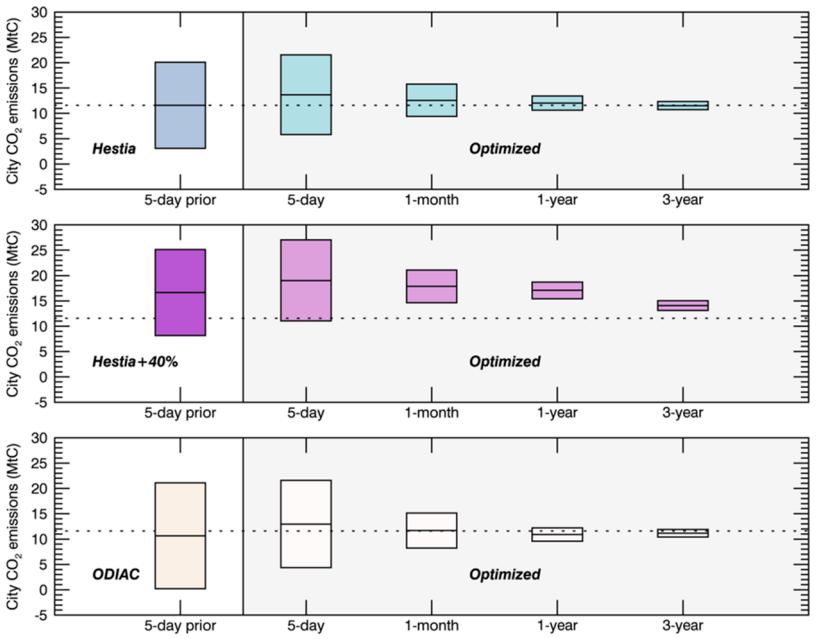

Figure 3. Whole-city daytime fossil fuel CO_2 emissions in MtC normalized over 3 years (September 2012−September 2015) for different *a priori* emissions (5 day estimates) and optimized emissions for 5 day, monthly, annual, and 3 year estimates. The central lines represent the mean absolute differences from Hestia emissions and the box heights represent the associated uncertainties (posterior uncertainties after optimization). Hestia whole-city daytime emissions are represented by the dashed lines.

fluxes. During the three dormant seasons, optimized $FFCO_2$ emissions are slightly lower than the initial Hestia estimate (−2 to −3.2%). When exploring the two specific $FFCO_2$ emission sectors (mobile and stationary sources, cf. Figure 2), the atmospheric solution varies from no $FFCO_2$ flux adjustment to a solution in which mobile $FFCO_2$ emissions are increased slightly (+2%) and stationary sources are reduced (−6%). The atmospheric constraint for each sector starts to deviate from the initial Hestia emissions estimate when larger uncertainties are prescribed to the coal-fired power plant (from 15 to 30% deviation in both the mobile and stationary sectors). However, the higher confidence reported from hourly energy production statistics[39] and the degraded statistical metrics (lower goodness-to-the-fit) do not support prescribing larger uncertainties to the emissions from the power plant. Overall, the atmospheric and bottom-up constraints (fossil fuel emissions) lie within 3% of each other annually.

More importantly for $FFCO_2$ emissions mitigation policy, the absence of understanding of interannual variations in whole-city daytime emissions limits our ability to quantify the amount of information added by atmospheric observations. To evaluate the influence of the atmospheric constraint, we introduced a seasonally dependent bias to the bottom-up Hestia $FFCO_2$ flux constraint and computed the final mismatch at different time scales. For a 15% positive bias imposed on Hestia, the atmospheric constraint produces a median correction of −14.2%, removing nearly all of the introduced bias (Figure 2). About one-fifth of the 5 day emission estimates fall within 10% of the original Hestia values. The overall agreement between the two constraints is about 2.5% over the 3 years despite the additional bias introduced to the bottom-up constraint. Seasonally, the median of wintertime adjustments is about −16%, slightly overadjusted, whereas

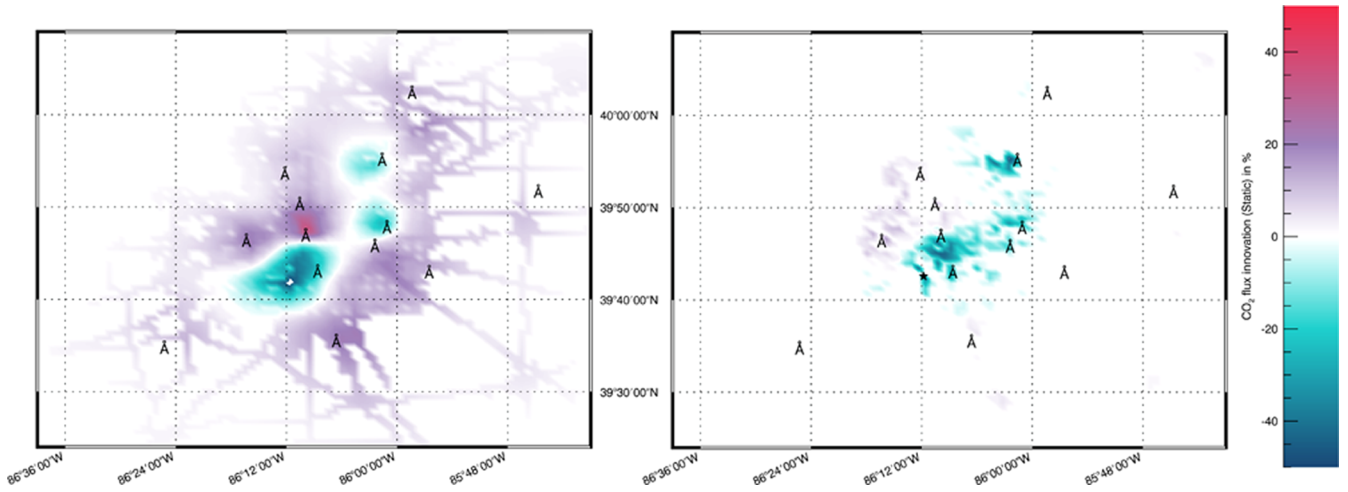

Figure 4. Maps of 3 year adjustments in CO_2 emissions (in %) after optimization aggregated from September 2012 to September 2015 for the mobile (left panel) and stationary (right panel) sectors with the measurement network (triangles). Hestia emissions were used as *a priori* emissions (first guess) in the CO_2/CO optimization system.

summertime adjustments are about −8%, lower than wintertime adjustments due to higher uncertainties in biogenic fluxes.

We further explore the importance of granularity from the Hestia bottom-up emission estimates in light of the fact that most cities across the globe have limited information related to the spatial distribution of their emissions. Here, we use alternative emissions at a coarser resolution derived from spatially disaggregated national emissions using nightlight satellite data, the Open-source Data Inventory for Anthropogenic CO_2 (ODIAC)[34] (SI Appendix, Section 4). We find that the optimized daytime emissions (11.2 MtC) agree within 3% of the Hestia-based optimized emissions (11.5 MtC) (Figure 3, upper panel). When assimilating only CO_2 mixing ratios, the convergence diminishes slightly with an $FFCO_2$ underestimation of 6% over the 3 years, suggesting that CO mixing ratios increase the contribution of fossil fuel emissions compared to biogenic fluxes. However, the spatial adjustments required in the optimization differ significantly between the Hestia-optimized and the ODIAC-optimized cases (SI Appendix, Figure S7). Uncertainties associated with the ODIAC-optimized emissions decrease significantly from 8 MtC over the 3 years to less than 1 MtC, indicating a much higher confidence in the combined (inverse) estimates. In the absence of robust temporal error correlations over long time scales, we assumed that 5 day estimates are uncorrelated which, if not true, might affect the prior (increasing uncertainties) but also the error reduction (decreasing uncertainties). We also note here that sectoral information is not available for ODIAC, an important limitation for establishing mitigation policies. When high granularity from Hestia is available for the bottom-up flux constraint, the optimization indicates small space-specific adjustments (Figure 4, lower right). For example, negative emission adjustments are applied to the southwestern and eastern parts of the city for both sectors while an increase in emissions is observed in the downtown area and in the city outskirts for traffic emissions (cf. Figure 4, left panel). The measurement sites located near the negative flux adjustments sample primarily highway emissions which might be the cause of the spatial adjustment. The spatial adjustments in static sources suggest an overall decrease in the southeastern quadrant of the city where the population grew only by less than 5% (Census population data), whereas the northern and western parts of the city experienced the largest population increase (>15% over 2000–2016). The spatial differences might indicate that CO_2 emissions have increased more rapidly in fast-developing areas (mostly suburban) since 2002 (i.e., the Hestia reference year for the spatial distribution).

The quality and amount of information collected to produce the Hestia CO_2 emissions for Indianapolis, IN is unprecedented, challenging to reproduce for many cities across the world. Assuming that lower-quality emission products will be available elsewhere, we explore the influence of the atmospheric constraint by increasing further the bias applied to the initial bottom-up flux constraint in Figure 5. As noted

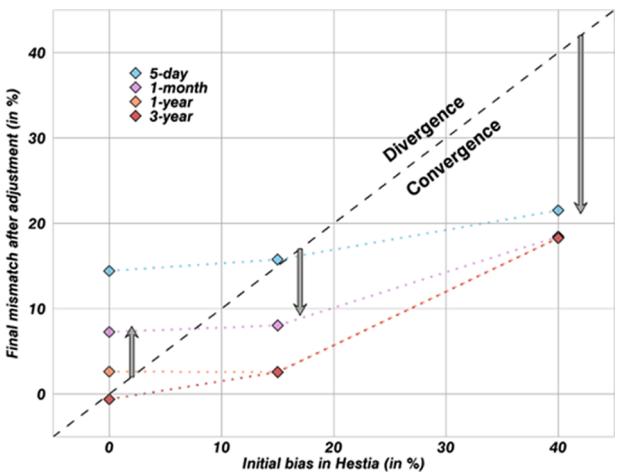

Figure 5. Mismatch between adjusted emissions and Hestia estimates as a function of introduced bias in bottom-up flux constraint. Mismatches are represented by the mean absolute differences (in %) aggregated over 5 day, monthly, annual, and 3 year time periods.

previously, an initial bias of less than 20% resulted in a final flux adjustment of less than 2.5% over 3 years and within 16% for 5 day averages. Annually, the initial bottom-up constraint and the final optimized flux remain within 3% of each other. Based on this result, satellite-driven bottom-up estimates such as ODIAC provide sufficient precision to produce whole-city

CO_2 emissions at policy-relevant levels over most US cities.[32] However, when the initial mismatch is larger (+40%), the atmospheric constraint is unable to adjust the optimized flux to reach similar levels of agreement (now within 18%), even at monthly, annual, or 3 year time scales. More than half of the initial bias has been removed, but the disagreement (18%) remains larger than policy-relevant levels (defined as 2% per year, or 6% over 3 years). Other recent studies suggested city-scale uncertainties of about 60% (mean relative differences) over Poland and the northeastern US, too large to converge using satellite-derived emission prior in the current optimization system.[7,40] We note here that even with a non-biased initial bottom-up flux constraint, 5 day and monthly estimates deviate by 14 and 8%, respectively, which may relate to errors in the atmospheric constraint or actual variability not accounted for in the Hestia bottom-up constraint. For large initial biases (here +40%), about 85% of the 5 day periods show an improvement and a fifth of the 5 day periods show a close agreement (within 10%), with a median bias adjustment of −24% (Figure S8, SI Appendix). Despite the temporal aggregation over 1−3 years, a severely biased bottom-up constraint (here +40%) will not be fully corrected by the atmospheric constraint.

Atmospheric assimilation approaches, like any other optimization procedure, rely on specified uncertainties, both in the fluxes, boundary conditions, and in the transport model. With an improved atmospheric model built on meteorological data assimilation (Supporting Information), flux and boundary condition uncertainties likely remain the two dominant sources of uncertainty. Because biogenic fluxes are more uncertain than $FFCO_2$ emissions, even at the whole-city scale, the potential for convergence of $FFCO_2$ is severely impacted by biosphere carbon exchange uncertainties during the growing season (summer adjustments are 50% weaker than wintertime adjustments in our bias-recovery experiments). Attribution to $FFCO_2$ fluxes or biospheric carbon exchange in the optimization scheme is mostly dictated by the presence of distinct spatial patterns, as human activities and biosphere overlap only in suburban areas in Indianapolis, and by the assimilation of CO mixing ratios assuming negligible contribution from the biosphere. Here, optimized summer emissions between June and August remain in agreement with Hestia, and despite the presence of large biogenic fluxes, no incorrect adjustment has been observed (Figure 1). We performed an additional experiment by introducing a 15% bias in biogenic fluxes while preserving their spatial and temporal distributions. The atmospheric optimization propagates part of the bias to $FFCO_2$ emissions due to misattribution of the signals, causing an underestimation of 7.5% at annual and 3-year time scales. Hence, careful estimation of biogenic fluxes is a prerequisite to urban $FFCO_2$ emission quantification, similar to findings for Boston, MA.[17] For higher granularities (e.g., sectoral biases), we performed sector-specific experiments (SI Appendix, Figure S9). Sectoral perturbations of 15% were added to one of the two sectors. After optimization, biased traffic emissions were adjusted down (−12%), an overall improvement, counterbalanced by a decrease in stationary sources (−6%), a misattribution of the bias from the traffic sector. When perturbing the stationary sector, stationary sources are adjusted by 8% (reducing by half the bias) but mobile sources decreased by 12%, a similar misattribution of the bias from stationary sources. Sectoral attribution depends on the relative weights of uncertainties and deviates from the original solution despite the assimilation of sector-specific tracers such as CO mixing ratios.[27] We conclude here that additional tracers are required beyond CO to improve the attribution of sectoral biases.

In the light of our results, high-resolution bottom-up CO_2 emission products similar to Hestia seem to provide necessary information for policy makers to establish mitigation strategies at the urban scale. However, important aspects to which atmospheric data provide complementary and timely information must be taken into account. Most metropolitan areas have no established policies for data access and often lack expertise to construct reliable emission products. In addition, fast-growing urban areas will require constant monitoring of sources as technologies and activities evolve rapidly while bottom-up products are delayed by data collection and changes in emission factors. Emission products like Hestia require a considerable amount of time and effort to compile and produce emission maps at high spatiotemporal resolution. Within the Indianapolis city boundaries, the direct emissions (Scope 1) from the city's self-reported inventory (SRI) are 35% lower than our optimal emissions estimate for the year 2013.[41] Both non-CO_2 and indirect emissions were subtracted from the reported community emissions for this comparison. At the sectoral level, traffic and industries CO_2 emissions are lower by more than 40% and the utility sector (electricity production) is lower by 25%. An accurate portrayal of the emitting landscape is essential to prioritize mitigation choices. The implementation of effective climate policies on robust estimates is essential to reducing gradually CO_2 emissions. Considering the mitigation strategy of the city, the 2025 target of the Thrive Indianapolis report aims at a reduction of 20% compared to the 2016 reported emissions, i.e., a change of less than 2% per year on average over the 10 year period. However, the systematic differences between the city SRI emissions and our optimal estimate remain too large to confirm the decrease in emissions by 2025 using the typical SRI methodology.

Due to limited information on soil organic carbon and above ground biomass, emission products currently exclude green carbon, while carbon storage is potentially a key player in offsetting carbon emissions from cities.[42] Most importantly, bottom-up products have no guarantee of continuity over multiple years due to continuously evolving activities, a fundamental limitation in this approach when characterizing interannual variations on the order of a few percent. By assimilating atmospheric measurements into the information system, timely information with long-term calibration will help preserve the monitoring of all activities across the urban area and over years, including the green fraction, in combination with available bottom-up products despite potential gaps in activity data or emission factors. Current and future deployments of eddy-covariance flux towers will provide critical measurements to calibrate and evaluate biogenic fluxes over urban areas. The density and locations of atmospheric stations remain dependent on the city characteristics (e.g., urban area extent, urban development, topographical features), but high density is desirable to map each district and sector of the area.[19] Technological advances in instrument techniques is key to increase the number of sensors produced at lower costs,[43] hence to reach sufficient coverage to provide, in association with bottom-up emission products, policy-relevant information for local governments and the private sector over most major metropolitan areas of the world.

This atmosphere monitoring/bottom-up integrated system over Indianapolis sheds light on the potential of atmospheric and information science to help quantify city $FFCO_2$ emissions but also to identify possible discrepancies at fine scales, i.e., seasonally, sectorally, and interannually. Constraining urban emissions at short time scales (submonthly) remain challenging with largely unexplained variations in 5 day means and a limited convergence at a high temporal frequency, as revealed by perturbation experiments. However, annual biases of whole-city emissions at the current level of mitigation policies seem attainable by combining granular bottom-up data products with atmospheric measurement approaches. Typical emission trends observed during the last decade are at the order of a few percent a year (2% per year achieved for the top third of C40 cities). With an optimal detection potential of 3−4% of the net $FFCO_2$ emissions annually (about 2% over 3 years), less than 5 years will be enough to confirm the reported changes with relatively high confidence. Similarly, fast-developing economies with annual growth rates in GDP of 6% or higher, assuming CO_2 emissions will grow at a similar rate, become even more measurable than trends in current reduction policy trends, measurable in less than 3 years. Going beyond the whole-city trends, decision makers will require higher granularity spatially and/or sectorally, to most efficiently target reduction measures. The current system includes CO mixing ratios to support the deconvolution of atmospheric signals into sectoral contributions.[27] Convergence in the stationary sources is weaker (converging from 40 to 29%) compared to mobile sources (from 40 to 23%), and 5 day standard deviation of sectoral emissions is about 26%, similar to whole-city emissions. This result suggests that whole-city sectoral changes are more difficult to capture with an atmosphere-only approach. Spatial attribution remains informative despite its sensitivity to uncertain error structures and will still require additional methodological development and improved uncertainty estimation before achieving the full potential of the integrated atmospheric/bottom-up optimization systems.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c00343.

> The observations and the inverse modeling systems are described in details, with complementary sensitivity experiments (PDF)

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Notes

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■ ADDITIONAL NOTES

[a] https://treaties.un.org/doc/Treaties/2016/02/20160215%2006-03%20PM/Ch_XXVII-7-d.pdf.
[b] https://www.c40.org/.
[c] 2017 US Census Bureau.
[d] www.carma.org; last access 27 March 2015.

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