



REVIEW

10.1002/2014EF000255

Special Section:

Urbanization, carbon cycle,
and climate change

Key Points:

- Large carbon fluxes and rapid change make cities key carbon cycle elements
- Cities represent ideal interdisciplinary carbon cycle process laboratories
- Sustained campaigns in representative cities will transform urban carbon science

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Citation:

Hutyra, L. R., R. Duren, K. R. Gurney, N. Grimm, E. A. Kort, E. Larson, and G. Shrestha (2014), Urbanization and the carbon cycle: Current capabilities and research outlook from the natural sciences perspective, *Earth's Future*, 2, 473–495, doi:10.1002/2014EF000255.

Received 23 MAY 2014

Accepted 7 AUG 2014

Accepted article online 13 AUG 2014

Published online 30 OCT 2014

Urbanization and the carbon cycle: Current capabilities and research outlook from the natural sciences perspective

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Abstract This paper explores the urban carbon cycle from the natural sciences perspective, identifying key knowledge gaps and priority areas for future research. The combination of large, concentrated carbon fluxes and rapid change makes cities key elements of the carbon cycle and offers the potential for them to serve as “first responders” for climate action. Estimates of urban-scale carbon fluxes are significantly more uncertain than at larger spatial scales, in part because past studies have mostly avoided local/urban scales where the mix of anthropogenic and natural fluxes is complex and difficult to observationally isolate. To develop effective emission reduction policies, we need to understand emission sources and how they may be changing. Such improved quantification and understanding of underlying processes at the urban scale will not only provide policy-relevant information and improve the understanding of urban dynamics and future scenarios, but will also translate into better global-scale anthropogenic flux estimates, and advance our understanding of carbon cycle and climate feedbacks across multiple scales. Understanding the relationship between urbanization and urban carbon flows requires intellectual integration with research communities beyond the natural sciences. Cities can serve as interdisciplinary process laboratories that are sufficiently constrained in both spatial and governance scale to support truly integrated research by the natural sciences, social sciences, and engineering. A thoughtfully crafted science research agenda that is grounded in sustained, dense observations relevant to estimating urban carbon fluxes and their controlling processes and is focused on a statistically significant sample of cities will advance our understanding of the carbon cycle.

1. Motivation

The relationship between urbanization, urban areas, and the carbon cycle has received increased attention from the natural science community in recent years driven by several intersecting interests and priorities. Foremost among these is the recognition that urban areas represent the dominant source of energy-related CO₂ emissions and a significant share of CH₄ emissions, proportions that are expected to climb as the global urban extent and the urban population grow disproportionately in the coming decades [Seto *et al.*, 2012a; United Nations Department of Economic and Social Affairs (UNDESA), 2012]. Furthermore, urbanization constitutes an important land-use and land-cover change process, with impacts on both terrestrial and aquatic carbon pools. The combination of large, concentrated carbon fluxes and rapid change in pools makes cities a large and dynamic element of the global carbon cycle. Between 2002 and 2011, global fossil fuel and cement production emissions averaged 8.3 ± 0.4 PgC yr⁻¹ [Le Quéré *et al.*, 2013], with over 70% of fossil fuel CO₂ emissions attributable to urban areas [Energy Information Administration (EIA), 2013]. Annual urban CO₂ emissions are more than double the net terrestrial or ocean carbon sinks [Le Quéré *et al.*, 2013]. Understanding and quantifying carbon flows in cities offers a powerful lens into urban ecosystems and provides a compact metric of urban sustainability [Bettencourt *et al.*, 2007; Fragkias *et al.*, 2013].

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The interest in urban carbon flows is additionally motivated by climate policy emerging at the local level responding, in part, to limited international and national policy progress [Rosenzweig *et al.*, 2010]. For example, over 1000 mayors have signed the U.S. Mayors Climate Protection Agreement, which commits them to meeting or exceeding the Kyoto Protocol reductions within their cities. Even if national and international policy progresses more rapidly than in the past, cities remain critical participants in the implementation of climate policy because the urban landscape is where the majority of industry operates, consumers live, and power is consumed. It is at the municipal scale that knowledge about local mitigation options, costs, and opportunities is the greatest [Betsill and Bulkeley, 2006; Fleming and Webber, 2004; Dhakal and Shrestha, 2010; Rosenzweig *et al.*, 2010; Salon *et al.*, 2010; Marcotullio *et al.*, 2014].

The recent attention to urban climate policy, in turn, has placed growing emphasis on understanding and quantifying current urban carbon flows and their potential responses to policy. Quantifying baseline emissions, planning emission mitigation efforts, and assessing progress toward climate mitigation targets all require improved monitoring, reporting, and verification (MRV) at the urban scale [Vine and Sathaye, 1999; Schakenbach *et al.*, 2006; Lutsey and Sperling, 2008; NRC, 2010]. Many developed megacities around the world are working together to pursue stabilization policies with greenhouse gas (GHG) inventories used by local governments, regulators, and business being a primary mechanism for assessing progress [C40, 2011]. In addition to implementing policies to reduce fossil fuel emissions, many cities have developed urban greening initiatives that seek to offset urban emissions through increasing uptake and storage by vegetation. Rigorous quantification and monitoring are required to ensure success of all mitigation efforts. Simple, first-order emission estimations based on population and economic characteristics are not accurate enough for our policy needs.

Interest among natural scientists in the urban domain also comes from researchers working at the global scale. For many years, the global carbon cycle science community has been studying the uncertainty associated with feedbacks between global climate change and carbon uptake on the land and in the oceans. Within that context, anthropogenic emissions, particularly the fossil fuel combustion component of those emissions (FFCO₂), are typically used as a near-certain boundary condition in assessing total carbon budgets. Assuming FFCO₂ emissions are known, other components of the carbon cycle can be studied by difference rather than requiring direct measurement [Gurney *et al.*, 2007]. For example, atmospheric CO₂ inversion and assimilation studies rely on an accurate quantification of fossil fuel CO₂ emissions [Enting, 2002]. It has been shown that errors in the location, timing, or magnitude of fossil fuel CO₂ fluxes can be aliased (transferred) onto the remaining flux components of carbon inversion studies [Gurney *et al.*, 2005]. Though traditionally considered well quantified, the accuracy of the FFCO₂ flux is sensitive to the space and time scales needed for analysis. At the global and annual scales, uncertainties have traditionally been estimated at 6%–10% [Marland and Rotty, 1984]. However, the global-scale uncertainty of the FFCO₂ has been increasing with time, driven by the increase in emissions from developing countries. With uncertainty at the global/national scale growing, uncertainty at the spatial scale of individual cities is far larger or unknown—estimates of 50%–100% error in the emissions estimates are not uncommon in the literature [NRC, 2010; Rayner *et al.*, 2010]. Furthermore, CH₄ uncertainties are large at all spatial scales [Miller *et al.*, 2012; Kirschke *et al.*, 2013; Brandt *et al.*, 2014]. The current uncertainties associated with urban emissions typically exceed emission reduction goals, making verification of emission reduction goals very challenging.

The large uncertainty associated with urban carbon emissions is due, in large measure, to the relative dearth of systematic, comprehensive, scientifically driven data collection at the urban scale. Emissions monitoring data that have been collected for regulatory or economic purposes are often incomplete or rarely checked against scientific standards and procedures. For example, mandatory CO₂ monitoring at large power plants in the United States shows little bias when examined in the national-aggregate scale but shows considerable uncertainty when examined on an individual facility basis [Ackerman and Sundquist, 2008]. In other sectors, such as transportation, which can be the largest sectoral CO₂ emitter in a city, there are missing or inconsistent data [Gately *et al.*, 2013]. Similarly, the locations and magnitudes of fugitive emissions from urban point sources, such as leaks in natural gas infrastructure, are largely unknown [Phillips *et al.*, 2013; Jackson *et al.*, 2014]. These problems result in significant scale gaps (between facility- and national-level emissions) and sectoral gaps in understanding. Additionally, the lack of information about the temporal characteristics of urban carbon emissions represents another difficulty

in establishing causal links between controlling activities and behaviors and emissions for key sectors. For example, although weekly cycles in emissions have been previously described [Cervery and Balling, 1998; Nassar et al., 2013], the consequences of these types of fine temporal variations for scaled-up annual emissions have only recently been explored [Peylin et al., 2011].

All of these intersecting interests highlight an urgent need to connect the social, political, economic, engineering, and physical dimensions of urban carbon flows and the urbanization process itself. Though admittedly complex, urban areas offer an ideal “process laboratory” for this purpose because they contain the complete suite of anthropogenic-driving activities and biogenic fluxes. They are the focal point for many political decisions that affect carbon, as well as being home to many products of human innovations and activity, including the built environment itself. Improved quantification and understanding of underlying processes at the urban scale will not only provide policy-relevant information to urban practitioners and improve the understanding of urban dynamics and future scenarios, but will also translate into better global-scale anthropogenic flux estimates, and thereby advance our understanding of carbon cycle and climate feedbacks at multiple scales. However, a complete understanding of the relationship between urbanization and urban carbon flows requires intellectual integration with research communities beyond the natural sciences.

To that end, this paper reviews the urban carbon cycle from a natural sciences perspective, with a focus on our current understanding and capability to quantify and model urban carbon flows through the land, water, and air. We then explore those elements of research where collaborations with engineers and social scientists are necessary to enrich our complete understanding of carbon and urban areas. Figure 1 provides a conceptualization of the urban carbon cycle, highlighting transboundary carbon movements, drivers, and flows. The scientific community must provide better constraints on the processes, pools, and

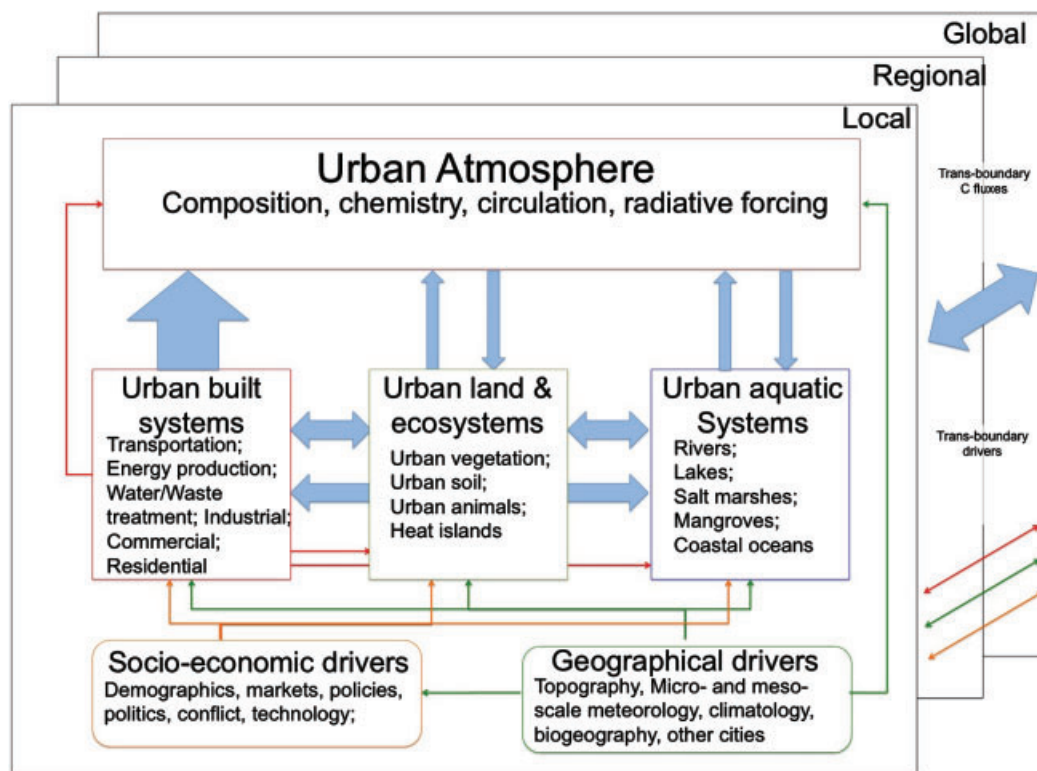


Figure 1. Urban carbon budget schematic showing key urban carbon reservoirs and processes (colored boxes), carbon emission and removal fluxes (blue block arrows), major drivers (rounded rectangles), and examples of process linkages (colored thin arrows). Examples of carbon pools and key emission and removal processes of particular importance are indicated within each reservoir box. The outer boxes acknowledge the relationship between the local-scale carbon budget of a given city and surrounding region and, ultimately, global through transboundary (lateral) carbon fluxes as well as interconnected drivers (socioeconomic, geographical, and built systems).

fluxes in order for us to add robust numeric values to this diagram. To that end, addressing the following scientific questions will fundamentally advance our understanding:

1. What are the urban anthropogenic carbon fluxes? How are these fluxes and associated carbon pools changing in time and space? How are they likely to change in the future?
2. What are the primary causes for discrepancies between research-grade and regulatory or “self-reported” emission inventories? Can we reconcile “top-down” and “bottom-up” approaches to quantifying FFCO₂ emissions?
3. Can we attribute fluxes to their underlying processes and resolve emissions in space and time?
4. How are these emissions manifested across cities and what are their sensitivities to the many controlling factors in different urban environments: geography, topography, climate, ecosystem type, socioeconomics, and engineering/technological factors? Are there emerging urban typologies for carbon emissions?
5. How do we apply natural science information on urban carbon flows to support and assess climate change policy options and assess efficacy?

2. Current Research Approaches to Understanding Urban Carbon

Research undertaken by the natural sciences into urban carbon flows has been focused primarily on quantification of carbon pools (e.g., fossil fuels, urban biota, aquatic systems, carbon-containing materials, and the atmosphere) and the fluxes between pools. Of particular interest to both carbon cycle research and the decision-support needs is the flux between the urban surface and the atmosphere. Flux estimation typically relies upon direct flux observations, atmospheric monitoring of CO₂, CH₄ and related species, and model/data approaches that rely on local data such as energy statistics, traffic modeling, and stack monitoring. Often, these methods are combined in various mixtures depending on the available observing systems, the relative magnitude of the carbon pools/fluxes in a given urban area, and the resources available.

The approaches to flux characterization pursued by the natural sciences can be broadly categorized as “top-down” and “bottom-up.” Top-down efforts can be further subdivided into methods that rely primarily on atmospheric CO₂ mixing ratio measurements and those that downscale larger-scale carbon budgets using various space/time proxies (e.g., remote sensing and socio-demographics). Bottom-up approaches start with data (energy statistics, land parcel data, and traffic monitoring) and models (building energy demand and travel demand modeling) on individual emitting entities—power plants, factories, and vehicles—and assemble the urban landscape emissions through integration. In practice, research has often availed of mixtures of both approaches. Note that, although not discussed in this paper, the term “top-down” can also refer to macroscale and mesoscale economic approaches for emission estimation [e.g., economic input-output tables; *Wright et al.*, 2011a; *Baynes and Wiedmann*, 2012].

Bottom-up approaches to urban carbon research have typically quantified carbon flows for a whole city or the census tract with sector- or fuel-specific information. For example, a number of studies have quantified fossil fuel CO₂ emissions at the scale of an entire county or city [*Baldasano et al.*, 1999; *Ngo and Pataki*, 2008; *Ramaswami et al.*, 2008; *Hillman and Ramaswami*, 2010; *Kennedy et al.*, 2010; *Parshall et al.*, 2010; *Sovacool and Brown*, 2010]. Other studies have attempted somewhat smaller spatial scales by quantifying emissions at the census tract or “community” level [*VandeWeghe and Kennedy*, 2007]. Most of this research has focused on urban areas within high-income countries where data are more readily available and methods are more harmonized. Much of this work has been motivated by improved understanding of urban “metabolism” or the relationship between urban form/function and energy flows (see *Chester et al.*, Positioning infrastructure and technologies for low-carbon urbanization, submitted to *Earth's Future*, 2014).

In addition to the academic literature, urban practitioners and nongovernmental organizations have engaged in carbon “footprinting” efforts as part of climate action plans or aspirational emission mitigation goals [*Wright et al.*, 2011a]. Though the methods and comprehensiveness of these individual city efforts are not consistent, they offer a diverse, though qualitative, illumination of the dynamics at play in urban carbon flows.

An approach to urban carbon flows unique to the natural science perspective has been the attempts in recent years to quantify emissions at the scale of individual buildings and street segments for whole

cities [Gurney *et al.*, 2012]. This is particularly important when attempting to link emissions to atmospheric concentration measurements within or near urban areas, to support MRV efforts [e.g., McKain *et al.*, 2012]. However, there is also potential for these fine-grained efforts to offer much more detailed and sectorally discrete information in support of urban-based mitigation policies [Schulz, 2010]. Fine-scale FFCO₂ estimates also offer a useful comparison point for satellite observations (see section 2.2.2).

Top-down approaches are categorized here as methods starting with atmospheric mixing ratio measurements used to infer fluxes through inversion of atmospheric transport and methods that downscale fluxes quantified at larger space/time scales using proxies. The approach based on atmospheric measurements has a relatively long history at larger space/time scales [Enting, 2002]. For example, atmospheric CO₂ model inversions were begun in the 1980s in an attempt to quantify large-scale fluxes of carbon between the land, ocean, and atmosphere with CO₂ concentration measurements as the primary observational constraint. Critical to this approach is simulation of atmospheric motions that link the observed constraint to the fluxes of interest. Scientists studying atmospheric chemistry and air quality have solved closely related estimation problems at the global scale over the past several decades through long-term, cross-calibrated measurements of multiple pollutant species [e.g., Prinn *et al.*, 2000; Brasseur *et al.*, 2003]. Translation of these approaches to the urban domain presents a number of challenges that are only in very recent years being overcome [McKain *et al.*, 2012; Bréon *et al.*, 2014]. In addition to the observational constraint associated with the flux of interest, atmospheric tracer transport models require meteorological observations at fine space/time scales to properly simulate atmospheric transport between the flux source and the location of atmospheric CO₂ measurements. Many of the transport processes at the urban scale are notoriously difficult to simulate due to small-scale turbulence, highly heterogeneous surface characteristics, and changes in the planetary boundary layer (PBL) height [Lauvaux *et al.*, 2009].

Downscaling flux estimates from larger space/time scales to the urban domain has used proxy information of many differing types. State- or national-level consumption or activity data have typically been downscaled using metrics such as urban extent, population, road density, and affluence, but these proxies can introduce biases in the estimates where the relationship between emissions and proxies breaks down or becomes nonlinear. The fundamental relationships between emissions and the proxies may also be geographically variable and statistically nonstationary.

Directly measured, long-term time series of emission fluxes from cities are scarce and the common carbon cycle tool of eddy covariance can be challenging to implement in urban areas due to urban structural heterogeneity [Grimmond *et al.*, 2002]. However, recent eddy flux measurements from an aircraft platform show promise in this regard [Cambaliza *et al.*, 2013]. Furthermore, land- and water-based carbon stocks and fluxes in and around urban areas have not been systematically measured, much less in conjunction with atmospheric observations and in the context of understanding carbon cycle dynamics in urban ecosystems. The discussion below is not intended to be a comprehensive treatment of carbon cycle science writ large; rather, we focus on key urban facets of the problem for which there is some data availability. Further, while linkages with climate change and air quality are clearly coupled to the urban carbon cycle, the scope of this discussion is largely limited to urban carbon flows.

2.1. Urban Fossil Fuel Emission Estimation

2.1.1. Space/Time Explicit

The capabilities associated with bottom-up efforts at urban carbon quantification have advanced dramatically in recent years. Numerous studies have quantified urban carbon budgets at whole city/annual scales and there exists rich literature in terms of quantity, methodological approach, and scope [e.g., Kennedy *et al.*, 2010; Chavez *et al.*, 2012; Hoesly *et al.*, 2012; Chester *et al.*, submitted manuscript, 2014]. The natural science community's interest in quantifying urban carbon emissions with space/time detail is driven by the importance of the atmospheric mixing ratio constraints and assessing how urban carbon flows are actualized in the atmosphere. Atmospheric measurements have two important attributes that make this linkage critical: (1) mixing ratios of CO₂ (and most GHGs) can be measured with considerable accuracy (well-established techniques can achieve accuracies of <1 part in 400) and (2) atmospheric mixing ratios provide an integral constraint due to atmospheric mixing [Weiss and Prinn, 2011]. This latter

attribute poses problems when interest turns to attribution of fluxes to specific elements of an emitting landscape because uptake and release processes occur simultaneously across the landscape. The separation of FFCO₂ emissions from CO₂ emitted by biotic activity can be achieved with carbon-14 (¹⁴C), but further separation of the fossil fuel signal into space or functional class (buildings versus roads) is challenging.

In order for atmospheric CO₂ measurements (¹²C or ¹⁴C) to be used as a constraint on flux estimates at the surface, those flux estimates must be correctly placed in space and time. The desired resolution of the underlying space/time fluxes is closely related to the atmospheric mixing ratio observational density, the complexity of the meteorological conditions/observations, and the adequacy of the atmospheric transport modeling capabilities.

The explanation for the relative dearth of fine-resolution efforts is likely a combination of the fact that interest and motivation in high-resolution work is very recent and that the data requirements have typically far exceeded availability in most urban areas. However, the availability, regularization, and connectedness of fine-scale data have grown tremendously in the last decade as cities make information such as land parcel data, socio-demographic data, traffic data, and pollution reporting publically available in digitized and common formats.

It is worth noting that the recent efforts focused on quantifying FFCO₂ emissions at suburban resolution are also drawing from urban planning and transportation research communities. For example, agent-based, microsimulation models have been developed, which attempt to simulate activities at an individual level (person, household, firm, or other agents) within the urban domain [Fissore *et al.*, 2011] and capture the interactions between land use and transportation [e.g., Waddell, 2002; Miller *et al.*, 2004]. Similarly, energy consumption and energy conservation policy measures have also generated high-resolution modeling and data systems research that focuses on residential and commercial buildings within a whole-city treatment [e.g., Shimoda *et al.*, 2007; Richardson *et al.*, 2010a]. Though historically not emphasizing GHG emissions, these communities have begun to apply planning tools to the problem of energy consumption and environmental impact, of which GHG emissions is a recent focus [e.g., Keirstead and Calderon, 2012].

2.1.2. Definitions, Modalities, and Frameworks

Quantifying urban carbon emissions as part of a system that includes atmospheric CO₂ monitoring has implications for the accounting framework adhered to. This is an important distinction between the space/time explicit quantification and the quantification that has been undertaken at coarser scales for policy considerations. Accounting frameworks for GHG emissions focus on how to assign responsibility for emissions and are often chosen to best match local interests or, more often, type or thoroughness of available data. The integration with atmospheric CO₂ measurement necessitates what has been variously called “production-based” accounting, “geographically-based” accounting, or “in-boundary” (IB) accounting [e.g., Chavez and Ramaswami, 2011; Wright *et al.*, 2011a; Ramaswami and Chavez, 2013]. IB accounting is also directly aligned with “scope 1” emissions following the WRI/WBCSD corporate GHG accounting schema [World Resources Institute/World Business Council on Sustainable Development (WRI/WBCSD), 2011]. IB emissions accounting quantifies all direct carbon emissions within the geographic boundaries of the urban area of study. It concerns itself with direct, physical fluxes of carbon between the land and atmosphere within a city and hence, does not include carbon embedded in goods/services, waste, and electricity produced outside the given urban area. IB accounting can be contrasted to consumption-based accounting which assigns emissions to consumers engaged in not only direct emitting activity but also indirect emitting activity such as the production of goods and services that give rise to emissions far from the consumption location [BSI, 2013].

A number of other characteristics are important when describing the distinction between the bottom-up emissions quantification pursued within the natural science community and “footprint” efforts intended to satisfy policy needs [e.g., Wright *et al.*, 2011b]. In order to maintain the fidelity of bottom-up emissions to adjustments as deemed necessary by an atmospheric observational constraint, bottom-up data products are sector, fuel, or even “process” specific. Sector categories are most commonly defined as industrial, residential, commercial/institutional, transportation (onroad, nonroad, air, and waterborne), and electricity production. It has been common practice to isolate municipal or government operations when

performing responsibility-based accounting. This is often considered within the commercial/institutional sector in the sector-based categorization.

“Process” refers to a more specific emission activity to which a singular emission factor or emission process can be applied. There are many different processes that emit carbon-containing compounds into the atmosphere and specifying which processes are considered is critical—not only to the accuracy of the emissions, but to the representativeness of emissions at a particular space/time scale. The most dominant and hence the most common process captured in urban carbon emissions research at high resolutions is the combustion of fossil fuels. Examples include emissions from gasoline internal combustion engines or bituminous coal boilers. However, other processes emit CO₂ via other carbon oxidation processes, such as cement manufacturing, fugitive emissions (e.g., evaporation of gasoline), and miscellaneous industrial processes (e.g., ammonia or aluminum production). Furthermore, combustion of other material, synthetic or organic, must also be considered. Indeed, the consideration of biogenic material is complicated as it typically necessitates consideration of the upstream CO₂ removal from the atmosphere to properly account for the net emissions. Similarly, net exchange of CO₂ with the biosphere outside of biotic material included in combustion is another process requiring careful accounting. Given the importance of quantifying the net exchange of atmospheric CO₂ with vegetation to CO₂ mixing ratios measured in the atmosphere, this category is critical (see section 2.4).

The spatial domain (or extent) considered by researchers varies, and typically this reflects the lack of a singular definition for what constitutes an “urban” area. Multiple data sources exist that define urban areas depending upon the needs and uses of the organizations or applications involved [see *Parshall et al.*, 2010; *Raciti et al.*, 2012]. These definitions can have a considerable impact on both the resulting emissions and their functional mixture. For example, inclusion of suburban areas can increase the need to consider the net exchange of carbon with vegetation, as suburban areas typically have higher vegetation cover than the urban cores [*Hutyra et al.*, 2011a]. With the growth of megacities and the agglomeration of previously separated urban areas into urban corridors (e.g., the Boston-Washington, DC megaregion), these domains can become larger than U.S. states and incorporate a wide array of carbon-emitting activities and large biological fluxes.

The spatial resolution of existing studies has also varied and is similarly driven by the questions posed and the available data. The finest scale attempted in the research literature is at the individual built structure and street segment (e.g., from intersection to intersection) level [*Shu and Lam*, 2011; *Gurney et al.*, 2012; *Gately et al.*, 2013].

From this finest resolution, a number of other resolutions are reflected in the literature. These generally follow definitions embedded within available data or defined along well-known and understood service territories. For example, land parcels, which can contain many buildings, are used when building footprint information is unavailable. Other candidates include zip codes, townships, neighborhoods (variously defined), and commercial zones. In onroad transportation, various spatial units are possible, the most common being the Travel Analysis Zone, which typically constitutes a homogeneous socio-economic portion of a city, ranging from fractions to a few square kilometers depending on population density.

An important consideration is the translation between “native” resolution, often comprising points, lines, and polygons, and regularized raster grids. The latter is essential to enable atmospheric transport modeling and the ultimate linkage to atmospheric CO₂ measurement. The choice of grid resolution is often dictated by the available modeling schemes and the density of meteorological data. Typical resolutions range from 200 to 1000 m (c.f. the engineering resolutions in *Chester et al.*, submitted manuscript, 2014).

Temporal resolution has traditionally kept to annual estimation with recent effort at monthly and even hourly time resolutions. Hourly resolution has a number of advantages. First, it allows one to resolve the day/night cycle in emissions—an advantage given that many atmospheric monitoring stations now sample air continuously and can potentially evaluate the emission time structure. Second, resolving nighttime emissions provides an additional opportunity to minimize the photosynthetic flux, a nuisance variable when attempting to isolate the fossil fuel CO₂ signal.

2.1.3. Case Studies

Quantifying bottom-up carbon emissions explicitly in space and time within the urban domain has generally focused on two subsectors: onroad transportation systems and buildings. This is not surprising as these two physical infrastructural entities and the activities they enable are large components of urban carbon emissions and tend to require the greatest level of effort to represent in fine space and time detail.

The transportation sector accounts for approximately one third of U.S. GHG emissions; onroad emissions account for 28% of U.S. GHGs [EPA, 2011]. Onroad transportation has been approached from a number of perspectives. Some approaches utilize the many decades of local and regional transportation planning where demand-oriented models are used to predict traffic volumes [e.g., *Nejadkoorki et al.*, 2008]. Other efforts have taken an activity-based approach whereby activity data such as vehicle miles traveled are combined with information on fleet composition and fuel economy [e.g., *Wang et al.*, 2009]; *Gately et al.* [2013] achieved annual emissions to the 1 km scale for Massachusetts with this type of approach. Finally, onroad emissions have been estimated with regression-based approaches. For example, *Bronfield et al.* [2012] found a combination of impervious surface area and road density offered a good model for the distribution of onroad CO₂ emissions. Similarly, *Shu and Lam* [2011] found best fit predictors using urban area, population density, and road density. Temporal distribution has been somewhat less studied for CO₂ emissions, but *Gurney et al.* [2012] used automatic traffic recorder to impute an hourly time structure. Detailed travel demand modeling has generated temporal emissions using travel scheduling [*Goulias et al.*, 2011]. Very high-frequency onroad speed, congestion, and volume information are also becoming available through tracking cell phone GPS signals; the linkage to FFCO₂ estimation is an active research area [*Herrera et al.*, 2010].

Quantifying the emissions from buildings at high resolution across the urban domain has typically relied on a combination of detailed bottom-up modeling and top-down spatial allocation. *VandeWeghe and Kennedy* [2007] allocated Toronto city-level annual emissions to census tracts in the residential sector based on energy payments. *Heiple and Sailor* [2008], by contrast, used building energy simulation modeling and geospatial databases to estimate residential and commercial building energy consumption down to the parcel level every hour. *Gurney et al.* [2012] built from the Heiple and Sailor approach and used the nonelectric share of building energy consumption at the individual building level to allocate county-level fuel-based CO₂ emission estimates on an hourly basis. A recent study obtained direct electricity consumption data associated with buildings in Los Angeles at the zip code+4 level, allowing for a data-driven estimate of building electricity consumption and the resulting consumption-based CO₂ emissions driven by this demand [*Pincetl et al.*, 2014]. Though not widespread, high-resolution energy consumption data holds tremendous promise for improved accuracy and knowledge generation at these scales.

The only study to date that has complemented the onroad and building sectors with all other FFCO₂ sources in a comprehensive high-resolution FFCO₂ emissions data product is the Hestia project [*Gurney et al.*, 2012]. The first Hestia product was developed for the city of Indianapolis, IN and included the addition of electricity production, nonroad emissions, aircraft, railroad, and industrial sector emissions for the year 2002 (Figure 2). Work is complete or underway on quantifying emissions at the same space/time scales for several other U.S. cities. The Hestia example uses several data sources and space/time conditioning data sets to estimate CO₂ and distribute the emissions in space and time. The result is spatial resolution that is a mixture of points, lines, and polygons at the hourly level.

2.2. Atmospheric Observations

Spatially resolved, bottom-up FFCO₂ data sets indicate that even small cities often present very intense, localized carbon fluxes of over 10 kgC m⁻² yr⁻¹ at spatial scales of 10 km² or less. Larger cities exhibit such fluxes in complex patterns over large areas (e.g., the LA megacity spans about 10,000 km²; Figure 3). These emissions lead to significant enhancements in the atmospheric mixing ratios of GHGs over the city relative to cleaner background air. The magnitude, distribution, and variability of these enhancements are driven both by local emission fluxes as well as meteorology and topography. For example, some cities exhibit “urban domes”—persistent and strong enhancements of CO₂, CH₄, CO, and other gases within the PBL above a city due to stable air associated with persistent inversion layers. Cities experiencing these

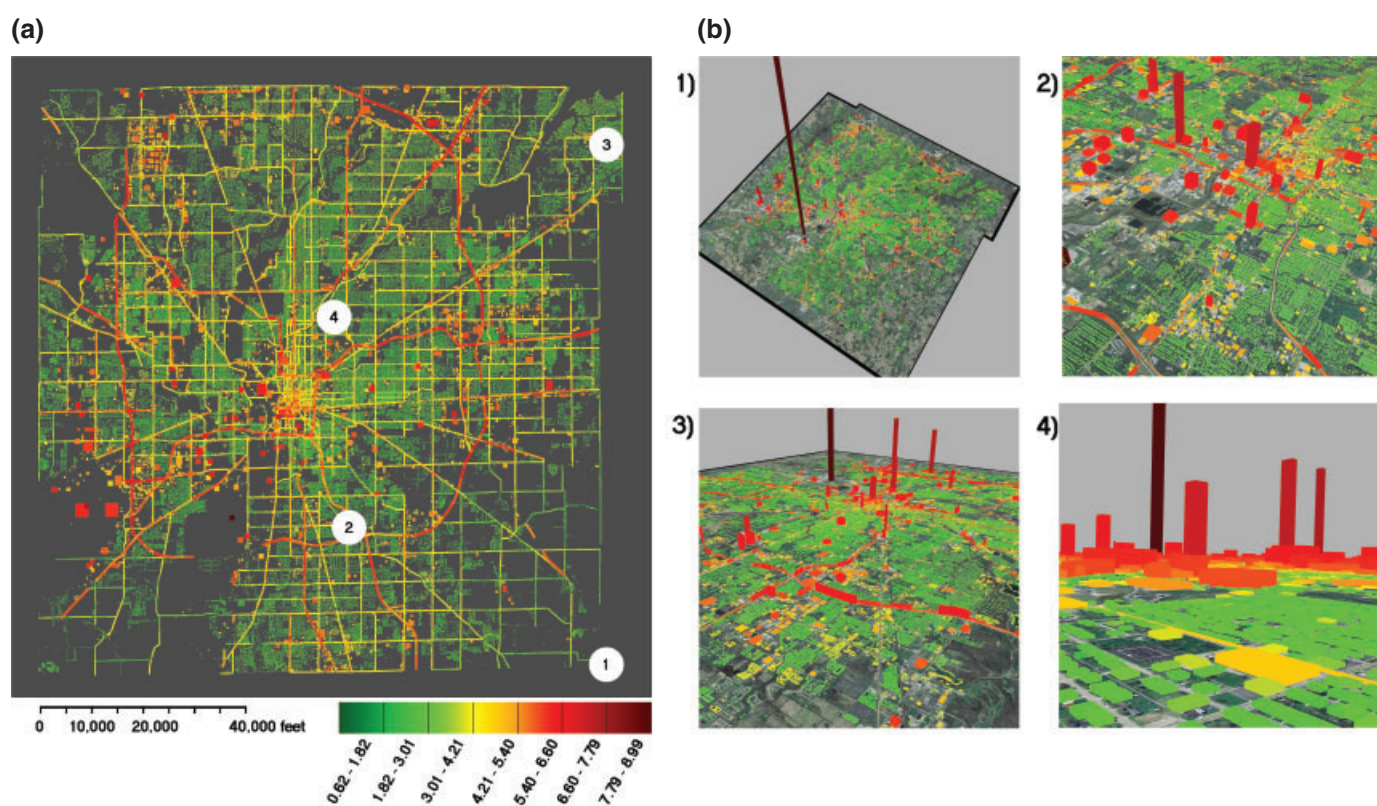


Figure 2. Hestia fossil fuel CO₂ emissions for Marion County, IN, for the year 2002: (a) top view with numbered zones and (b) blowups of the numbered zones. High-emitting roadways are typically high volume interstates and primary roads. Large vertical rectangles reflect power production and industrial sources. Color units: log₁₀ kgC yr⁻¹. Box height units: linear.

conditions, such as Los Angeles and Salt Lake City, routinely exhibit peak PBL CO₂ concentration enhancements of over 50 ppm relative to background air even though the net emission fluxes differ between larger and smaller cities [McKain *et al.*, 2012; Newman *et al.*, 2013]. “Urban plumes” manifest in cases where the enhancements associated with a city are dominated by prevailing winds—much like plumes from large point sources [Sparks and Toumi, 2010]. Cities in this category, such as Paris and Indianapolis, exhibit more diffuse and intermittent CO₂ enhancements than cities exhibiting urban dome conditions. In addition to strong spatial gradients, there are strong temporal variables that affect both local emission fluxes (e.g., energy and transportation demand cycles) and the evolution of airborne mixing ratios (e.g., diurnal changes in PBL height and different wind conditions). While FFCO₂ and CH₄ emissions from cities are typically significantly larger than nearby biogenic fluxes, the relative contributions vary by city and often exhibit strong seasonal dependencies (see section 2.4). Robust flux estimation depends on observational systems that address all of these variables. Urban CO₂ mixing ratio enhancements should not be assumed to be in steady state; FFCO₂ emissions respond to changes in human behavior.

Trace gases have been monitored in many cities for decades, although those efforts have mostly focused on reactive gases—criteria pollutants including ozone precursors, particulate matter, and other species important for air quality assessments. Existing air quality observational systems are, in many cases, not easily adaptable to studies of long-lived GHGs such as CO₂ and CH₄. Air quality monitoring is generally conducted at heights of 2–3 m above the ground and is designed to report threshold-crossing events for a given species. GHG monitoring, by contrast, is intended to estimate net emission fluxes for a much larger area and/or geolocate and quantify individual fugitive emission sources. These divergent objectives lead to different requirements on measurement precision, location, and modeling approaches. Measurement networks for GHGs and carbon fluxes have historically been located in rural areas to intentionally avoid cities as “confounding sources” of anthropogenic carbon—an appropriate strategy for quantifying biological carbon sources and sinks or background conditions. In recent years, top-down studies have

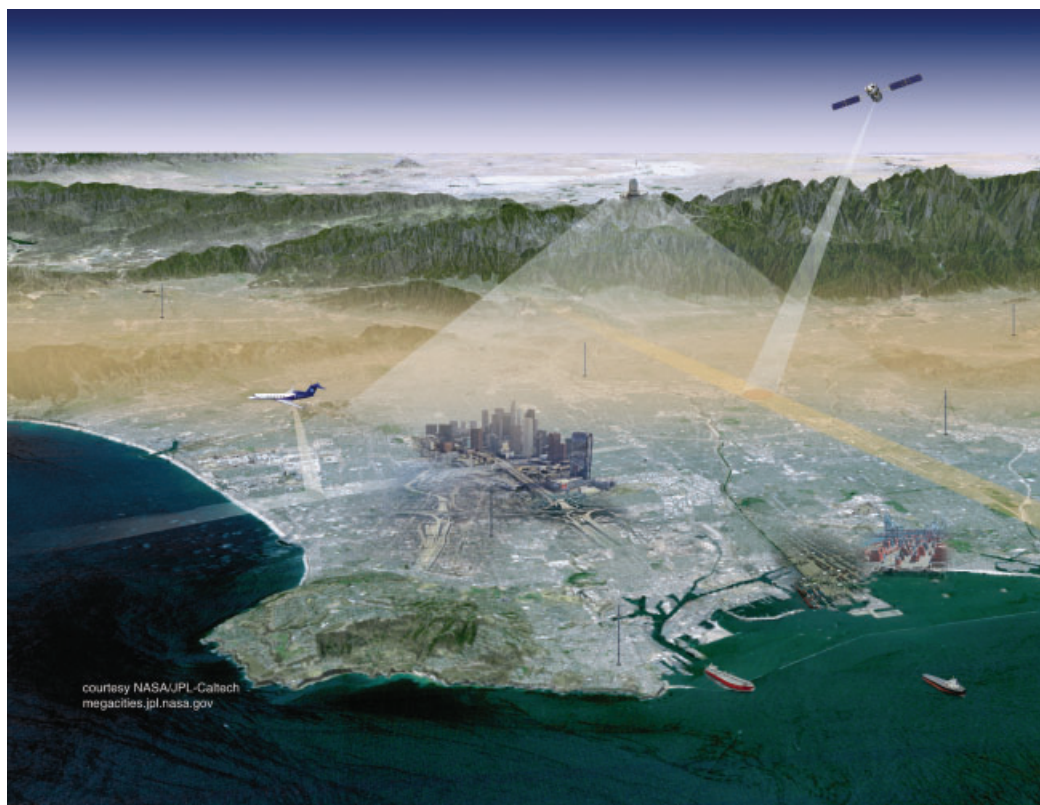


Figure 3. Conceptual view of a typical complex urban carbon environment and a tiered observational system. This illustration describes the Megacities Carbon Project in Los Angeles, CA, but is representative of elements shared with other urban carbon studies. In situ gas analyzers on towers and rooftops in and around the basin provide near-continuous, high-accuracy measurements of near-surface atmospheric mixing ratios of CO_2 , CH_4 , and CO . Aircraft and mobile laboratories provide infrequent but intensive measurements of mixing ratios, often focused on specific locations or emission sectors. Other surface-based instruments (not shown) measure winds and boundary layer height to validate the atmospheric transport models essential for translating mixing ratio data into carbon fluxes. Surface-based remote-sensing instruments provide daytime measurements of column-averaged mixing ratios. Satellites in polar orbit are beginning to provide column measurements of CO_2 and other species with sufficient sensitivity and resolution to characterize many cities around the world.

been piloted in selected cities to develop and test methods for using atmospheric mixing ratio observations of CO_2 , CH_4 , and other species to estimate emission fluxes, and also attribute relative contributions to specific sectors as an independent check on bottom-up estimates.

There are many options for observing the atmospheric abundances of CO_2 and CH_4 but they are all grounded in the application of spectroscopy to measure the strength of distinct infrared absorption lines of these gases. Some of the observational methods described here are also relevant to other GHGs including N_2O and fluorinated gases, but the focus of this work is on urban carbon (CO_2 and CH_4). Differences between methods primarily involve measurement location, specific spatiotemporal sampling schemes, and the sensing technology and algorithms used to retrieve mixing ratio estimates. Each method has strengths and limitations. Two major categories are described here: in situ sampling and remote sensing.

2.2.1. In Situ Observations of Atmospheric Mixing Ratios

In situ sampling involves “point measurements,” meaning air at a given location is directly measured—either quasi-continuously (e.g., integrating measurements every 1 s to a few minutes) by on-site gas analyzers or captured in flasks at some cadence (e.g., weekly) and stored for subsequent shipment and analysis in a laboratory. Flask sampling provides a powerful capability for laboratory instruments to retrieve mixing ratios for many atmospheric species in ways not practical for field instruments—including radioisotopes and other coemitted species. Radioisotope data in particular, while logistically challenging and expensive to collect, provide a unique ability to isolate the FFCO_2 sources given the depletion of radiocarbon (due to radioactive decay) compared to contributions of the contemporary biosphere. This

use of $^{14}\text{CO}_2$ data as a tracer for FFCO_2 has been demonstrated repeatedly from the ground and airborne platforms [Levin, 2003; Turnbull *et al.*, 2006, 2011; Graven *et al.*, 2009; Miller *et al.*, 2012].

While many in situ instruments are used to retrieve mixing ratios that are subsequently translated to flux estimates using inverse modeling and other analytical methods, another approach is to deploy instruments that measure fluxes directly. A flux tower combines measurements from a CO_2 gas analyzer and a colocated three-dimensional wind sensor with eddy-covariance techniques to directly estimate a flux [Baldocchi, 2003]. The eddy-covariance technique is used extensively in studying ecosystem fluxes [e.g., Wofsy *et al.*, 1993] with some application to urban domains [e.g., Grimmond *et al.*, 2002; Coutts *et al.*, 2007; Sparks and Toumi, 2010]. However, the effective footprint or sensitivity of such flux towers is typically very small, approximately 1 km in diameter. Further, urban locations can often violate the methodological assumptions for the eddy-covariance measurement. Most contemporary urban-scale carbon studies rely on inverse methods and atmospheric transport models, yielding footprints of receptor sites that are many kilometers in diameter—enabling coverage of an urban area with a reasonable number of instruments.

For urban studies, in situ gas analyzers and flask collection sites are often deployed in surface networks designed to sample both the local urban dome/plume, as well as relatively unpolluted background air, thus allowing estimates of the enhancement caused by the city's emissions [McKain *et al.*, 2012; Bréon *et al.*, 2014]. The sensitivity of measurements from a given site is strongly dependent on the horizontal and vertical location of the sampling inlet, sample frequency, proximity to terrain, prevailing meteorology, and local sources or sinks. Determining the sensitivity footprint of a given measurement site often requires applying Bayesian inverse analysis to assess the impact of prevailing winds and other conditions. For example, one such study for Los Angeles concluded that a minimum network of eight well-located sites should distinguish fluxes on 8 week time scales and 10 km spatial scales to within $12 \text{ gC m}^{-2} \text{ d}^{-1}$ or 10% of average peak fossil CO_2 flux [Kort *et al.*, 2012]. The same in situ gas analyzers used at surface sites can be optimized for faster sample rates and deployed on aircraft platforms to cover larger areas [Mays *et al.*, 2009; Cambaliza *et al.*, 2013].

Finally, in situ instruments are frequently deployed in automobiles to conduct street-level surveys in urban environments [e.g., Brondfield *et al.*, 2012; Phillips *et al.*, 2013; Jackson *et al.*, 2014]. Such campaigns are usually not amenable to estimating fluxes given the aforementioned limitations in sampling height and complexity of near-surface micrometeorology, although they can be useful in detecting persistent enhancements that might suggest a nearby point source.

2.2.2. Remote Sensing Observations of Atmospheric Mixing Ratios

Remote sensing-based carbon observations use atmospheric sounders to estimate a column-averaged dry air mole fraction of CO_2 , CH_4 , and CO (referred to as X_{CO_2} , X_{CH_4} , and X_{CO} , respectively). Column measurements have an advantage of being less sensitive to small-scale atmospheric motions that can impact estimates of fluxes derived from in situ point measurements [McKain *et al.*, 2012]. While some upcoming satellite instruments will use active (laser) methods, the established state-of-the-art involves passive solar spectroscopy. Surface-based, upward-looking instruments such as Fourier transform spectrometers (FTS) track the Sun to estimate X_{CO_2} , X_{CH_4} , X_{CO} and sometimes other species in the atmospheric column over a measurement site [Wunch *et al.*, 2011a]. Surface-based solar FTS instruments tend to provide higher signal to noise than satellite systems and are able to retrieve additional species. Solar FTS instruments have been deployed globally, such as NASA's Total Column Carbon Observing Network (TCCON), to support validation of satellite observations [Wunch *et al.*, 2011b]. Recently, TCCON sites have been installed in urban centers and adjacent exurban areas to support urban-background subtraction (e.g., at Caltech in the Los Angeles basin and in the nearby Mojave Desert) and smaller, more portable units are being tested in various cities for characterizing spatial gradients (e.g., across the Boston region).

Satellite instruments are now becoming available with near-surface sensitivity, precision, and spatial resolution sufficient to contribute to urban flux estimation. Examples include Japan's Greenhouse gas Observing Satellite (GOSAT) and NASA's Orbiting Carbon Observatory (OCO-2; launched in July 2014) [Miller *et al.*, 2007]. A recent study evaluated midday observations of X_{CO_2} from GOSAT collected over Los Angeles to

demonstrate that even nonoptimized space-based remote sensing can provide measurements of megacity CO₂ emissions [Kort *et al.*, 2012]. By taking the difference between observations over the megacity and nearby background measurements, Kort *et al.* observed robust, statistically significant enhancements in total X_{CO₂} of 3.2 ± 1.5 (1 σ) ppm. The Kort *et al.* [2012] results were consistent with ground-based X_{CO₂} observations [Wunch *et al.*, 2009], suggesting that these atmospheric enhancements can be exploited to track trends over time.

The strength of satellite observations is the ability to provide dense spatial sampling resulting in thousands of observations over short intervals that both provide spatial mapping over an area of interest and enable sample aggregation to significantly reduce random errors [Baker *et al.*, 2010]. The current generation of space-based CO₂ sounders was designed to study regional-to-continental scale natural carbon fluxes rather than cities and other point sources. Hence, they have significant gaps in spatial coverage and revisits for a given geolocation are often measured weeks apart. Potential future missions could frequently target cities, offering more precise flux estimates and diurnal temporal resolution [Duren and Miller, 2012; Key *et al.*, 2012; Buchwitz *et al.*, 2013].

2.3. Urban-Scale Analytical Frameworks (Top-Down Approaches)

To infer quantitative information on carbon exchange (fluxes) from observations of atmospheric mixing ratios, it is necessary to account for the influence of atmospheric transport. This is clearly illustrated in the example of Salt Lake City where observations of CO₂ near the surface exhibited a large diurnal variation primarily modulated by variation in the depth of the PBL [Strong *et al.*, 2011; McKain *et al.*, 2012]. In general, urban mixing ratios of GHGs reach a maximum at night when a shallow PBL exists, and a minimum midday when the deepest mixed layer and maximum ventilation occur. This contrasts with the diurnal variability of anthropogenic emissions, which typically reach maximum values during the day and minima at night. During the daytime hours, biogenic uptake also draws down atmospheric CO₂. Multiple techniques have been developed and exploited in order to account for atmospheric transport and link atmospheric observations to net carbon flux.

2.3.1. Transport Box Models

Box or mass-balance techniques present the simplest conceptual technique. This approach represents transport as a simple flow and uniform mixing process as opposed to numerical transport models, which attempt to simulate all transport process akin to weather models. In mass-balance approaches, a small number of boxes are typically considered, representing, for example, the urban zone and its surrounding region. The problem is thus simplified to a simple reservoir-flow construct—if one measures the change in water level (i.e., atmospheric mixing ratios) and observes the lateral flows in and out (upwind and downwind transport to and from the urban box), one can then quantify the input flow from the surface (net emissions into the box from the urban surface).

As part of the INFLUX study, an aircraft was used to sample the downwind plume of Indianapolis [Mays *et al.*, 2009]. Considering the enhanced CO₂ and CH₄ in the plume, PBL height, and wind speed, a net emission estimate could be derived. This approach is a powerful method to determine total net emissions, but is subject to both situational constraints and potential biases resulting from simplifying assumptions. It is necessary to have a rather isolated source which experiences steady winds to produce an urban plume—many urban regions do not meet these criteria. Potential variability in the PBL depth upwind and over the urban center has the potential to bias the results.

A simple multibox model has been constructed for Salt Lake City to investigate surface observations and disentangle the relative roles of transport, anthropogenic, and biogenic emissions [Strong *et al.*, 2011]. This approach facilitates a clear understanding of relative impacts from these three contributors, but fails to provide robust quantitative information on fluxes.

The box model concept has also been applied to satellite observations, considering the enhanced urban dome of CO₂ in Los Angeles relative to the surrounding region [Kort *et al.*, 2012]. By assessing the difference in column abundance over an urban region, changes in time in the urban dome and underlying emissions can be robustly detected. This method enables the tracking of annual time-scale emission changes, but relies on assumption of spatial stationarity of sources and transport on annual time scales. Quantification of contributing sectors or net flux is also not robustly retrievable.

2.3.2. Tracer-Tracer Correlation

A different approach to account for transport without explicit representation leverages the knowledge that conservative (on the time frame of interest) tracers experience the same atmospheric transport processes. With this knowledge, if one knows emissions of gas X with greater certainty than gas Y, and gas X and Y are either coemitted or have experienced sufficient mixing prior to measurement, the observed correlation Y:X can then be scaled by the relative molecular weights and the prior emissions of X to derive the emissions of Y. This technique provides a powerful mechanism to account for the actual transport experienced by the observed air mass and thus provides a simple approach to quantify net emissions of a gas. The technique is limited in that good knowledge of emissions of a gas is required, and error in that representation will directly transfer into gas Y, and collocation or sufficient mixing is required. Isolating geographic location or particular sector sources is not possible with this approach. This method has been applied with great success to Los Angeles in quantifying methane emissions [Wunch *et al.*, 2009; Hsu *et al.*, 2010; Wennberg *et al.*, 2012], where CO emissions are substantially better quantified than methane. A similar approach has been used to estimate changes in emissions in Beijing during the Olympic games, both from the ground [Wang *et al.*, 2009] and from space [Worden *et al.*, 2012]. Space-based estimates leveraging this approach have now been extended to many global urban centers [Silva *et al.*, 2013].

2.3.3. High-Resolution Mesoscale Modeling

In high-resolution mesoscale modeling atmospheric transport is explicitly simulated, and thus a quantitative link can be established between atmospheric mixing ratio observations and surface fluxes. In attempting to fully simulate atmospheric dynamics, this approach provides a powerful platform from which specific questions may be posed, including quantifying net carbon flux and spatiotemporally resolved carbon flux. Mesoscale modeling can, and has been, used to design optimal observational networks [Lauvaux *et al.*, 2009; Kort *et al.*, 2013]. Meteorological parameters have been optimized for studies within specific cities, demonstrating the importance of proper representation of the urban canopy [Nehrkorn *et al.*, 2013]. These methods have also been applied to quantify emissions and quantitatively track changes in emissions with time [McKain *et al.*, 2012; Brioude *et al.*, 2013]. These approaches provide a powerful tool for many urban carbon investigations, but require robust fine-scale estimates of FFCO₂ and biological fluxes as a model prior to estimate. Proper optimization for each urban environment presents a challenge, and adequate resolution simulations present computational challenges. Additionally, quantitatively addressing all contributing sources of errors presents a challenge.

2.4. Urban Biotic C Exchange

Until recently, urban vegetation and soils have not been considered significant within the carbon cycle research. However, biogenic exchange within cities can significantly influence local atmospheric mixing ratios [Day *et al.*, 2002; Coutts *et al.*, 2007; Vesala *et al.*, 2008; Briber *et al.*, 2013]. Further, the wide array of urban greening initiatives underway has the potential to strengthen the urban biogenic fluxes share of total urban carbon flows within cities.

The construction of urban areas results in widespread ecosystem modification, dramatically altering land cover and the flows of carbon in and around urbanizing regions. Land-cover changes associated with urbanization typically decrease carbon storage [Imhoff *et al.*, 2004; Hutyra *et al.*, 2011b; Seto *et al.*, 2012a], alter biogeochemical cycles [Kaye *et al.*, 2006; Pataki *et al.*, 2006; Grimm *et al.*, 2008] and hydrologic regimes [Walsh *et al.*, 2005; Pickett *et al.*, 2011], and influence micrometeorology and regional weather patterns [Oke, 1982; Zhang *et al.*, 2004; Zhou *et al.*, 2011]. Although urban areas are the major centers for energy consumption and the subsequent emissions of CO₂, they do also sequester some of the very same emissions they produce in urban soils and plant biomass [Imhoff *et al.*, 2004; McPherson *et al.*, 2005; Golubiewski, 2006; Kaye *et al.*, 2008; Raciti *et al.*, 2012; Briber *et al.*, 2013]. Urban vegetation has also been credited in aiding local carbon mitigation strategies [Nowak and Crane, 2002; McPherson *et al.*, 2005], but the urban FFCO₂ emissions typically dwarf local biogenic uptake [Pataki *et al.*, 2011; Briber *et al.*, 2013; Figure 4]. Nonetheless, the diurnal cycle of urban photosynthesis and respiration does significantly influence atmospheric mixing ratios of CO₂; photosynthesis draws down atmospheric CO₂ during the daytime when FFCO₂ is maximized [Briber *et al.*, 2013]. Potential urban carbon sinks are inherently limited due to a lack of available plant growing space and export of litter, but urban vegetation functions as a

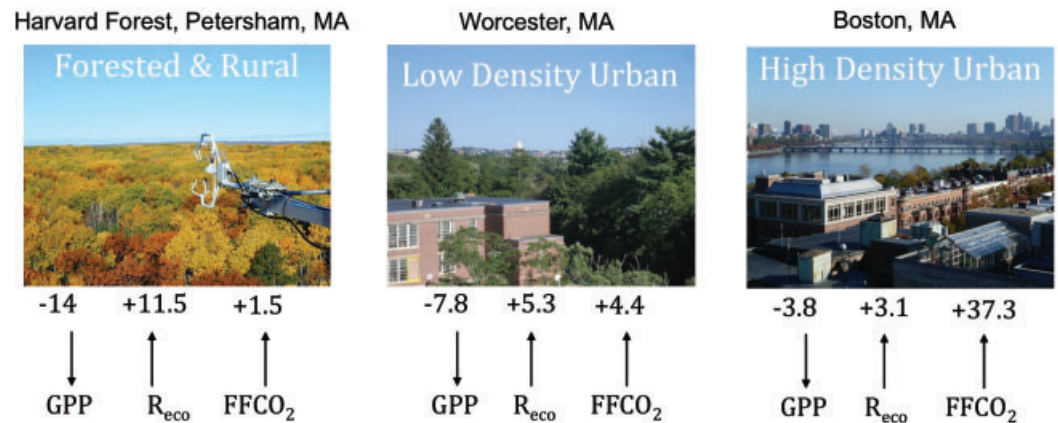


Figure 4. Sample gradient in carbon fluxes from biotic land fluxes and anthropogenic emissions [$\text{MgC ha}^{-1} \text{ yr}^{-1}$; after *Briber et al.*, 2013]. Gross primary productivity (GPP) represents biological uptake through photosynthetic process; ecosystem respiration (R_{eco}) represents carbon release through autotrophic and heterotrophic process, and fossil fuel emissions were extracted from *Gurney et al.* [2009]. Negative fluxes denote uptake of carbon by the land; positive fluxes denote release of carbon to the atmosphere.

vital component of cities providing aesthetic, economic, and ecological value to urban dwellers [*Nowak and Crane*, 2002].

Urban vegetation can constitute a significant portion of land cover within the urban mosaic, with proportions in major U.S. cities ranging from 10% to 30% of urban land area [*Nowak et al.*, 2001]. However, “urban” is often an inconsistently defined land cover. For example, *Raciti et al.* [2012] compared three commonly used urban definitions and found that vegetation carbon stock density estimates ranged from 37 ± 7 to $66 \pm 8 \text{ MgC ha}^{-1}$ for the urban portions of the Boston metropolitan area. *Hutyra et al.* [2011a] found an average of $89 \pm 22 \text{ MgC ha}^{-1}$ in vegetation within the Seattle Metropolitan Statistical Area lowlands, a carbon density comparable to intact temperate forests. In a follow-up study, they found that the carbon lost to the atmosphere due to clearing of land for expanding urban development accounted for nearly 15% of local fossil fuel emissions, making it an important, but overlooked, term in the regional C budget [*Hutyra et al.*, 2011b]. The influence of biological land fluxes on atmospheric CO_2 mixing ratios will vary as a function of the urban definition/extent, vegetation types, and biome.

The aggregate effects of urbanization (including changing land-cover characteristics, land-use patterns, impervious surface fractions, urban heat islands (UHIs), extended growing seasons, atmospheric pollution, management activities, etc.) on land-atmosphere exchange processes remain poorly understood despite decades of study on specific aspects of urbanization [*Pouyat et al.*, 2006; *Canadell et al.*, 2007; *Churkina et al.*, 2010; *Hutyra et al.*, 2011a; *Pickett et al.*, 2011]. Interactions between urbanization and ecosystem carbon exchange are complicated by competing positive and negative feedbacks that vary across different regions and change over time. *Imhoff et al.* [2004] estimated that urbanization reduced net primary production (NPP) by 0.04 PgC yr^{-1} in the United States, but the uncertainty associated with this estimate is very high due to incomplete data and understanding of feedbacks. *Imhoff et al.* [2004] also show that through localized warming, “urban heat islands” can extend the growing season in colder regions and increase winter NPP. Using MODIS, *Zhang et al.* [2004] demonstrated that the timing of both leaf emergence and fall senescence is significantly influenced by UHIs in and around urban centers in the eastern United States. Specifically, they found that the growing season increased by as much as 15 days in areas affected by UHIs. Moreover, the signature of the changes in leaf phenology due to UHIs extends well beyond the footprint of urban land use [*Buyantuyev and Wu*, 2012]. Changes in growing season lengths have been widely linked to enhancement in uptake of CO_2 [e.g., *Richardson et al.*, 2010b]. Urban vegetation also often grows in favorable light conditions with open-grown tree crowns and higher diffuse light fractions. Additionally, recent research has shown that reductions in wind speed within urban areas may decrease stress on plants, increasing the productivity [*Bang et al.*, 2010].

Urbanization may also increase NPP in resource-limited regions by increasing nutrient availability through nitrogen (N) deposition [*Rao et al.*, 2014] and through increased water availability in arid areas [*Buyantuyev and Wu*, 2009; *Zhang et al.*, 2013]. Cities geographically concentrate activities such as transportation, food

and energy consumption, and lawn management making urban areas a major global source of reactive, gaseous forms of N [NO_x and NH_3 ; Vitousek *et al.*, 1997; Galloway *et al.*, 2003; Hall *et al.*, 2011]. However, elevated concentrations of NO_x and tropospheric O_3 can potentially offset stimulated rates of primary productivity [Reich, 1987]. Extensive national-scale nitrogen deposition monitoring networks exist in the United States and Europe, but similar to the global CO_2 monitoring networks, sites are intentionally located away from urban areas and point sources of pollution in order to capture background regional trends. There have been recent discussions and attempts to expand monitoring into urban areas [e.g., Fenn *et al.*, 2003; Lohse *et al.*, 2008] to address some of the challenges discussed in this paper.

Most efforts to model urban biogenic carbon flows have not been spatially explicit or have relied on coarse-scale remote sensing imagery and relatively simple light-use efficiency models [e.g., Zhang *et al.*, 2008; Zhao *et al.*, 2012]. Recently, maps of urban biomass at very high resolution have been produced [Davies *et al.*, 2011; Raciti *et al.*, 2014], and these data will facilitate improved modeling of urban productivity and net carbon fluxes as a function of the suite of changing urban environmental drivers.

An important additional component of the urban land and aquatic carbon cycle is black carbon (BC). Incomplete combustion of carbonaceous matter emits BC particles larger than $1 \mu\text{m}$ that may fail to become airborne or may otherwise quickly drop to the nearest surface. Precipitation and runoff events may transfer such particles accumulated on the soil surface to rivers and oceans [Clarke and Patterson, 1997; Masiello, 2004]. Alternatively, larger particles and charred materials may remain on the soil, near the point of production and deposition. With time, natural processes such as bioturbation and land development activities can drive the mixing and accumulation of such BC materials into the soil, promoting C sequestration and enhancing nutrient sorption [Shrestha *et al.*, 2010]. Approximately $12\text{--}24 \text{ TgC yr}^{-1}$ is produced as BC from fossil fuel combustion [Penner *et al.*, 1998]. Correlating urbanization history with fossil fuel-derived BC stored in soils [Liu *et al.*, 2011] could be used for assessing the long-term urbanization impacts. Biomass burning produces $50\text{--}260 \text{ TgC yr}^{-1}$ as BC of which approximately 80% is fire residue [Kuhlbusch and Crutzen, 1996], a significant contribution in the context of rising urbanization with biomass-based energy systems in developing nations. Large uncertainties in urban BC emissions and deposition estimates remain, largely due to bottom-up approaches that often do not correctly reflect recent socioeconomic changes in emerging economies [Rawlins *et al.*, 2008]. The first top-down, global-scale estimation of BC emissions used data from both column aerosol absorption optical depth and surface concentrations from global and regional networks (Kalman filters) to constrain a fully coupled climate-aerosol-urban model, deriving an optimized estimate of BC emissions as $17.8 \pm 5.6 \text{ TgC yr}^{-1}$ [Cohen and Wang, 2014]. Given the recalcitrance of BC in soil, it is worthy of additional consideration in accounting for urban carbon stocks and fluxes [Masiello and Druffel, 1998].

Taken as a whole, vegetation within cities is likely to significantly influence the urban carbon cycle on both long and short time scales. The overall contributions will vary with the biogeography and the urban form. In many respects, vegetation growing in urban environments foreshadows broad-scale global environmental changes with increased local CO_2 , temperature, and reactive N deposition, longer growing seasons, and significant land-use changes, offering a powerful “process laboratory” for studying the broader carbon cycle.

2.5. Urban Aquatic C Fluxes

Cities are likely to play a significant role in the fate of aquatic and coastal carbon. In 1990, approximately 44% of the global population lived within 150 km of a coast, and the majority within 100 km of a perennial river [Small and Cohen, 2004]. Further, 13% of the global urban population live within the low elevation coastal zone ($<10 \text{ m asl}$), with lesser-developed countries having a greater proportion than Organization for Economic Co-operation and Development (OECD) countries [McGranahan *et al.*, 2007]. Aquatic and coastal ecosystems within and adjacent to urban areas may contribute significantly to urban C flows, both as a means of transport for lateral flows and as hotspots of biogeochemical activity.

From a global perspective, inland waters and coastal areas are important components of the carbon cycle [Cole *et al.*, 2007; Battin *et al.*, 2009; Tranvik *et al.*, 2009; Raymond *et al.*, 2013]. Aquatic ecosystems export significant amounts of both inorganic and organic carbon from terrestrial sources to recipient systems and are hotspots [McClain *et al.*, 2003] of carbon transformation. The majority of large rivers are net sources of

CO₂ to the atmosphere [Cole and Caraco, 2001], while coastal ecosystems such as mangroves, salt marshes, and seagrass beds potentially sequester carbon (colloquially known as “blue carbon”) at rates significantly higher than forests [Mcleod *et al.*, 2011]. However, many global carbon models exclude fluxes from inland waters due to the difficulties associated with estimating the extent of these ecosystems, the portion of the flux that has already been accounted for in the terrestrial sink, the partial pressure of CO₂, and gas exchange rates between water and the atmosphere [Battin *et al.*, 2009]. Blue carbon sequestration is also frequently not included in estimates of global carbon sinks, which tend to divide the world into solely terrestrial or ocean compartments. Coastal ecosystems are difficult to map globally, and reported rates of burial by ecosystem type are highly variable [Mcleod *et al.*, 2011]. It has been proposed that incorporation of these aquatic fluxes, including outgassing to the atmosphere, storage in coastal biomass, and burial in sediments, may help reconcile the discrepancies between bottom-up inventories and top-down inversion estimates of CO₂ sinks [Battin *et al.*, 2009; Regnier *et al.*, 2013].

In addition to the difficulties mentioned above, the direct and indirect impacts of anthropogenic activities add further uncertainty to our understanding of C dynamics in both inland waters and coastal ecosystems. Erosion, nutrient additions, altered thermal and hydrologic regimes from agricultural activities, land-cover change, discharge of sewage water, and outright destruction of riverine and coastal ecosystems are just some of the anthropogenic activities that influence C flux [Walsh *et al.*, 2005; Mcleod *et al.*, 2011]. Regnier *et al.* [2013] estimated that anthropogenic perturbations have enhanced the flux of carbon to inland waters by as much as 1.0 PgC yr⁻¹, with 0.4 PgC yr⁻¹ then emitted to the atmosphere, and 0.5 PgC yr⁻¹ sequestered in sediments in freshwater systems, and only increasing the input to the ocean by 0.1 PgC yr⁻¹. Although Regnier *et al.* [2013] acknowledge low confidence for several of their flux estimates, they are some of the only values available. No such estimates are yet available for urban aquatic fluxes of carbon that are specifically urban.

The evaluation of anthropogenic drivers of C fluxes generally does not differentiate between urban and nonurban (e.g., agricultural and mining) activities. However, the type and intensity of activity may be very different within cities than outside. At the present time, global estimates of CO₂ fluxes from inland waters are not spatially explicit at a resolution that allows for disentangling anthropogenic impacts as they occur along the hydrologic flow path. Therefore, it is difficult to separate the indirect effects that cities have on aquatic carbon fluxes, resulting from teleconnections to nonurban ecosystems [Seto *et al.*, 2012b], from the direct influence of urban activities on aquatic ecosystems within and adjacent to urban areas. The aforementioned analysis of anthropogenic perturbation of aquatic carbon fluxes by Regnier *et al.* [2013] suggests that the most direct urban contributions stem from inputs to inland waters of sewage and soil C, potentially increasing sequestration [Lee *et al.*, 2006]. However, ambient urban conditions and the location of urban areas along the hydrologic flow path could have a significant effect on emissions from aquatic and coastal ecosystems as well. Urbanization is also frequently the cause of the destruction and resulting C loss for mangroves [Alongi, 2002], wetlands [Ehrenfeld, 2000; Dewan and Yamaguchi, 2009; Mitsch and Hernandez, 2013], and streams [Walsh *et al.*, 2005].

Many aquatic and wetland ecologists have conducted case studies to examine one or more components of the carbon cycle in urban aquatic and coastal ecosystems, evaluating, for example, the quality of dissolved organic matter in storm water runoff from urban areas [McElmurry *et al.*, 2014], or using stable carbon and nitrogen ratios to separate agricultural from urban wastewater C inputs to coastal sediment cores [Vaalgamaa *et al.*, 2013]. No study, to date, has evaluated the sum of all impacts in an urban area to determine the net loss or gain of carbon. Additional case studies, urban comparisons that vary both in development intensity and physiographic context, and improvements in data quality and resolution will be vital for assessing the net contribution of aquatic C fluxes to urban carbon budgets. These improvements are especially pressing given the hotspots of urban expansion to 2030 identified by Seto *et al.* [2012a], the majority of which occur on coasts, river corridors, lake shores, or small islands.

3. Conclusions and Future Research Needs

Urbanization is a major driver of global environmental change, and cities are key components of the global carbon cycle due to the combination of large, concentrated carbon fluxes and rapid change. These characteristics give cities significant leverage to act as “first responders” and influence the trajectory of

anthropogenic climate change [Rosenzweig *et al.*, 2010]. Unfortunately, as this article documents, the current uncertainties and paucity of data regarding urban carbon fluxes and biogeochemical and socio-economic processes that control those fluxes hamper our understanding and thus limit our ability to evaluate the efficacy of emissions mitigation efforts. The dearth of urban information is an artifact of historical priorities in carbon cycle research and the realities of observational limitations. Past efforts have primarily focused on characterizing carbon sources and sinks at regional to global scales, avoiding local and urban scales where the mix of anthropogenic and natural fluxes is more complex and difficult to observationally isolate. However, recent advances in observing techniques have brought the urban spatial scale within reach and offer the opportunity to create scientifically credible carbon monitoring systems to verify fluxes and therefore advance effective mitigation policies.

It is critical for such carbon monitoring systems to account for both CO₂ and CH₄. Recognizing this, there are key distinctions in our current understanding of urban emissions of these gases. Most notably, as discussed above, “bottom-up” estimates of CO₂ emissions can leverage documented information on emission activities such as traffic and fuel consumption to produce reasonable first estimates. These “bottom-up” estimates can then be combined and compared with “top-down” estimates to provide optimized CO₂ flux estimates. In the case of CH₄, urban emissions do not linearly track with census or economic drivers, and exhibit difficult to predict spatially heterogeneous structure—dominant urban sources for CH₄ are natural gas pipeline leaks, landfills, and industrial flaring/venting. A direct consequence is that urban methane emission bottom-up estimates contain much greater uncertainty with no clear path to improvement in the near term. In the case of methane, “top-down” methods founded in atmospheric observations provide the only current path toward quantification of urban emissions.

Urban carbon studies have increased in recent years with diverse motivations ranging from urban ecology research to testing methods for independently verifying GHG emission inventories. However, these studies are still quite nascent, have been somewhat ad hoc thus far, and were primarily driven by the research interests of individual scientists or small research collaborations rather than a formal research priority with federal science agency leadership. For example, there has not yet been the equivalent of an urban study of comparable scale to the North American Carbon Program's Mid-Continent Intensive experiment, a comprehensive campaign to close the carbon budget of a Midwestern rural region using top-down and bottom-up methods [Schuh *et al.*, 2013], not to mention an effort that attempts to integrate the natural sciences, social sciences, and engineering factors relevant to carbon cycling. Additionally, current urban carbon studies are limited to a small number of cities that likely do not capture the range of urban typology needed to make robust inferences about the broader distribution (Table 1). As a result, important and yet incomplete progress has been made on the urban carbon front in developing methodological capabilities and applying them to resolving space-time variations in carbon fluxes or developing appropriate emission reduction strategies.

We suggest that cities represent ideal “process laboratories” in that they contain a complete suite of processes that control the large and complex anthropogenic and biogenic carbon fluxes. Cities can serve as interdisciplinary test beds that are sufficiently constrained in both spatial and governance scale to support truly integrated research by the natural sciences, social sciences, and engineering—all motivated by a common sense of importance and urgency in improving understanding of urban carbon flows. The research gaps identified throughout this paper can be closed through a thoughtfully crafted urban carbon science research agenda that is grounded in sustained, dense observations of atmospheric, land, and aquatic variables relevant to estimating carbon fluxes and their controlling processes from a statistically significant sample of cities. The definition of well-posed research questions and an urban carbon typology, followed by design and implementation of intensive and sustained urban campaigns in representative cities, should lead to advances in urban carbon understanding similar to that seen over the past decade of global carbon cycle science.

3.1. Linkages to the Social Sciences and Engineering Communities

Companion papers by Chester *et al.* (submitted manuscript, 2014) and Marcotullio *et al.* [2014] provide thorough reviews and discussion of the urban carbon cycle from engineering and social science perspectives, respectively.

Table 1. Representative List of Contemporary Urban Carbon Experiments

Study Type	Example Projects	Urban Population (Thousands of People)	Domain Size (km ²)	Likely Emission Trends	Signature of Airborne Enhancement	Dominant Emission Sectors	Local Terrestrial Biosphere	Local Aquatic Systems
Sustained monitoring, process attribution, and prototype decision support for megacities	Megacities Carbon Project (LA basin)	18,000	25,000	Declining	Dome	Transport (CO ₂), energy (CH ₄)	Urban green spaces, agriculture, chaparral	Coastal
	CarboCount-City (Paris/IDF)	12,000	12,000	Declining	Plume	Transport, residential (CO ₂)	Forest	River
"Top-down" flux estimation methodological studies	INFLUX (Indianapolis)	840	3,600	Stable	Plume	Transport, electricity generation	Agriculture	River
	Boston study	4,641	10,000	Stable	Plume	Transport, residential (CO ₂)	Forest	River, coastal
	Salt Lake City study	1,000	12,000	Stable	Dome	Transport, residential (CO ₂)	Sparse	Lake
Urban ecology	Seattle MSA	3,500	15,000	Stable	Plume	Transport, residential (CO ₂)	Forest	River, coastal

Chester et al. (submitted manuscript, 2014) discuss urban infrastructure and technology and the challenges faced in widening the interdisciplinary perspective on cities and the built environment when assessing carbon emissions. Links between the natural science research on urban carbon, as outlined here, and the new research emerging from the engineering sciences on urban infrastructure and technology are critical for a more complete understanding of the drivers of the carbon pools and fluxes highlighted in this paper. FFCO₂ emissions are embedded within increasingly complex and interdependent built environment systems and our ability to progress from diagnosis to prognosis will depend upon our ability to link the observational and modeling systems outlined in this paper to the built environment. For example, quantification of bottom-up emissions explicitly in space and time must develop a more mechanistic link to the processes represented in urban infrastructure and the built environment, and how people interact with the built environment. A more mechanistic representation then makes the rich past and emerging work on technological options, socio-technological systems, and the mixed "hard" and "soft" engineering approaches available to scenario development that can represent the engineering drivers through to atmospheric realizations. Similarly, the trajectory of future emissions associated with different infrastructure systems can also be represented alongside immediate emissions to provide a more complete picture of the present and future impacts and offer a more informed policy process. Finally, this link builds the first bridge toward the more complete conceptualization where the natural science modeling and monitoring is linked to the more complex mix of the built environment and the socio-behavioral drivers.

Marcotullio et al. [2014] explore the relationships between the process of urbanization, socio-institutional systems, and the carbon cycle. The social science of urbanization has only recently integrated energy use and subsequent FFCO₂ as important outcomes of patterns of urban development, governance, and social dynamics. Studies examining the relationship between socio-institutional systems, energy use, and FFCO₂ emissions have thus far produced consensus at only the broadest levels (e.g., urban wealth and urban energy use are correlated) and debate continues around the directionality and mechanisms underlying such relationships. Social science research examining the relationship between urbanization and the carbon cycle is nascent, appropriate data are only recently available, and the relationships are inherently complex. This argues for interdisciplinary collaborations among the sciences. The way societies organize now and into the future is a vast challenge facing humanity and generates uncertainty for estimates of future FFCO₂ emissions. We need to advance our understanding of how bioclimatic context, urban lifestyles, human behaviors, economies, demographics, and governance can lower the impact of urbanization on the carbon cycle and reduce FFCO₂ emissions.

Acknowledgments

LR Hutyra is supported in this research by NSF CAREER award DEB-1149471 and NASA Interdisciplinary Science award NNX12AM82G. R Duren's research was done at the Jet Propulsion Laboratory, a division of the California Institute of Technology under contract to the National Aeronautics and Space Administration. KR Gurney is supported in this research by NSF CAREER award 0846358 and National Institute for Standards and Technology grant 104336. N Grimm was supported by NSF BTS1026865. The October 14–17, 2013 Workshop on Human-Carbon Interactions in Urban Systems sponsored by the Carbon Cycle Interagency Working Group (CCIWG) and US Carbon Cycle Program provided the platform for this research. The data presented in this review paper are synthesized from existing literature; the corresponding author can be contacted directly for additional information.

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