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Constraining sector-specific CO_2 and CH_4 emissions in the United States

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Abstract. This review paper explores recent efforts to estimate state and national-scale carbon dioxide (CO_2) and methane (CH_4) emissions from individual anthropogenic source sectors in the United States. Nearly all state and national climate change regulations in the US target specific source sectors, and detailed monitoring of individual sectors presents a greater challenge than monitoring total emissions. We particularly focus on opportunities to synthesize disparate types of information on emissions, including emissions inventory data and atmospheric greenhouse gas data.

We find that inventory estimates of sector-specific CO_2 emissions are sufficiently accurate for policy evaluation at national scale but that uncertainties increase at state and local levels. CH_4 emissions inventories are highly uncertain for all source sectors at all spatial scales, in part because of the complex, spatially-variable relationships between economic activity and CH_4 emissions. In contrast to inventory estimates, top-down estimates use measurements of atmospheric concentrations to infer emissions at the surface; these efforts have had little success identifying CO_2 emissions from anthropogenic sources but have successfully identified sector-specific CH_4 emissions in several opportunistic cases. We also describe a number of forward-looking opportunities that would aid efforts to estimate sector-specific emissions: fully combine existing top-down datasets, expand intensive aircraft measurement campaigns and measurements of secondary tracers, and improve the economic and demographic data (e.g., activity data) that drive emissions inventories. These steps would better synthesize inventory and top-down data to support sector-specific emissions reduction policies.

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1 Introduction

Government regulations of greenhouse gas (GHG) emissions have evolved rapidly in the past five years, particularly in the United States. For example, the US Environmental Protection Agency (EPA) recently announced CO_2 and CH_4 emissions regulations for numerous source sectors as part of the White House Climate Action Plan (Executive Office of the President, 2013). Several US states have also taken aggressive action on emissions, including Massachusetts (Massachusetts Executive Office of Energy and Environmental Affairs, 2015) and California (Air Resources Board, 2014), among others.

These policy actions require that scientists and government agencies quantify regional- and national-scale GHG emissions from specific source sectors. This review paper focuses on existing and evolving capabilities for the United States. The US has far greater resources to estimate emissions relative to many developing countries. Furthermore, GHG emissions regulations in the US are nascent relative to regulations in Europe (e.g., Prahl and Hofman, 2014), and the monitoring strategies discussed in this review could be developed in parallel with new regulations.

This focus on individual source sectors is important for supporting recent US GHG emissions policies. In this paper, we define a source sector as the total emissions from an industry, such as CO_2 from power plants, CH_4 from the oil and natural gas industries, or CH_4 emissions from landfills. Emissions from specific components of these industries are beyond the scope of this review (e.g., emissions from gas wells versus gas storage systems). Most national emissions regulations in the US target this sector level. For example, the US Clean Power Plan mandates a 32% decrease in power sector CO_2 emissions by 2030 relative to 2005 levels (In February, 2016, the Supreme Court stayed implementation pending a final court ruling.) (US EPA, 2015a). The EPA and National Highway Traffic Safety Administration have also extended and strengthened CO_2 emissions standards for cars and light trucks through 2025 (US EPA Office of Transportation and Air Quality, 2012). In addition to these measures, EPA has set several sector-specific CH_4 emissions targets. In August of 2015, EPA proposed a rule that would decrease CH_4 emissions from oil and gas operations by 40–45% relative to 2012 levels (US EPA, 2016b). Last but not least, the EPA announced proposed regulations for CH_4 emissions from landfills in August 2015 (US EPA, 2015b).

Emissions from these source sectors are important to quantify not only at national scale but also at the state level. US federal policies like the Clean Power Plan are implemented through plans devised by each state; each state has a different emissions reduction target, and each state can decide how to meet and monitor progress toward that target (US EPA, 2015a).

We examine sector-specific GHG estimates with an eye toward combining or assimilating multiple data streams. This review article is part of a special issue of the European Geophysical Union (EGU) journals that focuses on data assimilation and the use of multiple data streams to understand the carbon cycle. In this context, we explore opportunities to creatively synthesize both bottom-up emissions inventories and top-down atmospheric inverse modeling. Most government agencies estimate emissions using bottom-up inventories: quantify total emissions by estimating the total amount of some activity and the average emissions per unit of activity. Other efforts utilize top-down atmospheric inverse modeling: measure atmospheric GHG concentrations and use those measurements to infer the level and distribution of emissions at the Earth's surface. Given current policy needs, no single strategy (i.e., bottom-up or top-down) will likely be sufficient to evaluate GHG emissions from specific source sectors. In the future, scientists and government agencies will likely need to combine these approaches to robustly estimate sector-specific

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emissions – frameworks that can synergistically leverage the information content of bottom-up datasets, atmospheric GHG data, and top-down strategies. This review paper focuses on these opportunities.

Future efforts to synthesize these strategies will need to address two key tasks: estimate the total quantity of GHG emissions from each source type and detect changes or trends in emissions from that source type. From the standpoint of inverse modeling, the former problem is more challenging than estimating total emissions and requires separating the space-time patterns of one emissions source from the patterns of other sources. In the latter case, we not only need to estimate a trend in total emissions but also need to attribute that trend to a specific source sector(s). This challenge is complicated by changes in technology and changes in the spatial or temporal distribution of individual source sectors. For example, hydraulic fracturing and horizontal drilling have risen meteorically in the past decade (US Energy Information Administration, 2015). These operations utilize new equipment and operational practices, and the spatial distribution of drilling across the United States has changed during that time. These changes can complicate efforts to estimate trends in CH_4 emissions from the oil and gas industries; these emissions are literally a 'moving target.'

These challenges are further complicated by GHG fluxes from the biosphere, particularly in the case of CO_2. In many instances, anthropogenic emissions are also co-located with natural GHG fluxes or fluxes caused by human-caused disturbances to the landscape. These natural and anthropogenic emissions will be important to disaggregate from one another for sound policy evaluation. For example, a natural landscape disturbance and subsequent change in CO_2 fluxes could be mistaken for a trend in human-caused GHG emissions (or vice versa).

In this article, we explore these challenges from several perspectives. First, we discuss bottom-up inventory efforts. We then explore top-down strategies to estimate sector-specific emissions and the atmospheric datasets available to make both bottom-up and top-down estimates. Next, we highlight several new or novel approaches for estimating sector-specific emissions, and lastly, we close the review with a synthesis discussion of forward-looking opportunities for combining bottom-up and top-down strategies.

2 Bottom-up data

Bottom-up efforts typically use an accounting-type approach to estimate sector-specific emissions. The first step usually involves collecting activity data: a map or database of economic activity or behavior that leads to emissions. Examples include the amount of coal burned by power plants, the number of passenger cars and miles travelled, and the number of cows by location. A second step entails estimating a set of emissions factors (EFs) for each activity. EFs could include the CO_2 emissions per kg of coal burned or the average CO_2 emissions per mile travelled by passenger cars. The product of these two numbers provides a bottom-up estimate of emissions for a given source sector. State and national governments in the US use this strategy to construct official emissions estimates (e.g., California Air Resources Board, 2015; US EPA, 2016a). A number of academic and government efforts have produced bottom-up CO_2 and CH_4 emissions estimates at local/regional (e.g., Gately et al., 2013; Jeong et al., 2014; Lyon et al., 2015; California Air Resources Board, 2015), national (e.g., Petron et al., 2008; Gurney et al., 2009; Gately et al., 2015; US EPA, 2013; Environment and Climate Change Canada, 2016), and global scales

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(e.g., Rayner et al., 2010; Andres et al., 2011; Oda and Maksyutov, 2011; Olivier et al., 2014). In this section, we primarily discuss bottom-up data with an eye toward how this information can be combined with top-down strategies.

2.1 A prototypical example

We describe EPA's estimate of CO_2 emissions from coal-fired power plants as a prototypical example of how government agencies construct bottom-up inventory estimates. EPA describes the procedure that it uses to estimate CO_2 emissions in compliance with 2006 IPCC guidelines (US EPA, 2016c): first, the agency estimates activity data – coal use by source sector. EPA uses retail statistics from the electricity sector to estimate total consumption by each type of end user (e.g., residential, commercial, etc.). Second, EPA adjusts this activity data to account for non-combustion uses, double-counted emissions, and fuel exports/imports. For example, a coal gasification plant in North Dakota produces synthetic natural gas; this fuel is added to natural gas activity data and subtracted from the coal activity data. Third, EPA estimates the carbon content of the coal. EPA uses Energy Information Administration (EIA) estimates of carbon content by coal rank and state of origin (Hong and Slatick, 1994). EPA then computes the weighted average carbon content of coal by state of origin and estimates the end use of coal produced in each state (e.g., electricity, industry, etc.). The agency uses this procedure to estimate the average carbon content (and EF) for each end use sector in the United States (US EPA, 2016c).

IPCC guidelines also require a reference approach: an additional verification or consistency check against fuel production, imports, and exports (US EPA, 2016a). The new draft inventory then goes through expert review undertaken by a panel of technical experts. EPA revises its inventory estimate based upon this review and distributes the subsequent draft for public comment. At the conclusion of that process, EPA issues its finalized inventory estimate.

The approach outlined above is prototypical of many government inventories. More recently, a number of academic efforts have developed very different approaches that leverage novel data streams (e.g., satellite images of night lights) or that use gridded activity data, and these efforts are described in detail in the next section.

2.2 Recent bottom-up efforts

In the past ten years, inventory efforts have moved from coarse estimates that rely heavily on proxy activity data to spatially-resolved estimates that use specific activity data and EFs that are tailored to the heterogeneities in each emissions source.

A number of recent CO_2 inventory efforts have incorporated more comprehensive activity data or detailed EFs than previously available. At the regional scale, Gurney et al. (2012) and Gately et al. (2013) developed on-road CO_2 emissions estimates for Indianapolis and Massachusetts, respectively. The latter study reports emissions that are within 8.5% of Federal Highway Administration fuel consumption statistics but that differ from the commonly-used, global-scale EDGAR inventory by 22.8%. The authors explain that many global-scale efforts use road density as a proxy for vehicle emissions but argue that the relationship between road density and emissions is not constant. Two subsequent studies (McDonald et al., 2014; Gately et al., 2015) estimate on-road CO_2 emissions for the entire United States at spatial resolutions down to 1 km^2. McDonald et al. (2014) estimate emissions that differ from EDGAR by 20-80% at the municipal level, though the two inventories produce nearly identical national totals.

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At national scale, the VULCAN inventory (Gurney et al., 2009) is the most comprehensive academic effort to date. The inventory includes CO_2 emissions by sector at high spatial and temporal resolutions – 10km $\times$ 10km and sub-daily. Furthermore, the inventory uses more detailed activity data than government efforts. For example, the inventory identifies emissions from individual point sources, a contrast to EPA's estimate which reports only county-level point source totals. At the global scale, the EDGAR anthropogenic emissions inventory has moved from a $1^\circ \times 1^\circ$ lat/lon resolution to $0.1^\circ \times 0.1^\circ$ (Olivier et al., 2014). In a separate effort, Andres et al. (2011) estimated CO_2 emissions for 80 countries with a particular focus on estimating the seasonal cycle of CO_2 emissions.

A number of studies have also leveraged more rigorous activity data and EFs to estimate anthropogenic CH_4 emissions. Jeong et al. (2014) and Lyon et al. (2015) estimate oil and gas CH_4 emissions for California and the Barnett Shale region, respectively. Both find emissions that greatly exceed EPA's estimates. A relatively small fraction of emitters account for the majority of oil and gas emissions, and Lyon et al. (2015) argue that rigorous EFs capture this skewed distribution more effectively than those used by EPA. In addition to these oil and gas inventories, Owen and Silver (2015) compiled field studies of CH_4 emissions from agriculture (e.g., cows, sheep, and manure management). The authors explain that current emissions inventories use EFs from lab-based experiments, not field observations. These field observations imply much higher EFs, EFs that result in higher emissions that are more consistent with existing top-down estimates.

A number of additional studies also leverage novel inventory methodology or novel proxy datasets. For example, Oda and Maksyutov (2011) developed ODIAC (Open source Data Inventory of Anthropogenic CO_2 emission), a global, gridded CO_2 inventory constructed using a database of CO_2 point sources and remote sensing data of night lights. Rayner et al. (2010) and Asefi-Najafabady et al. (2014) developed a data assimilation framework known as FFDAS (Fossil Fuel Data Assimilation System). The authors use datasets like population density and economic activity as inputs into their model, constrain or fit their emissions model using nightlight data, and reported national emissions totals. Davis and Caldeira (2010) used a very different approach from any of the above studies. The authors build a CO_2 inventory based upon economic imports and exports and explore the idea of carbon 'leakage', the carbon emitted by one country to manufacture products that are then imported by another country. These studies do not provide emissions estimates for each individual source sector, but ODIAC and FFDAS do leverage novel datasets to separate out point sources (e.g., power plants) from non-point emissions. Overall, most of the above inventory efforts (except EDGAR) are the product of academic, not government, research.

EPA's GHG Reporting Program (GHGRP) represents an important advancement in government inventory efforts. EPA announced the GHGRP in 2009 and emissions reporting began in 2010 (US EPA, 2013). The GHGRP requires all entities that emit over 25000 metric tons of CO_2 equivalents to report their emissions to a national registry (US EPA, 2013). This reporting threshold is equivalent to the GHG emissions of 3439 homes or 5263 cars (US EPA, 2015c). The agricultural sector is excluded from this threshold and is not required to report its emissions. Despite this omission, EPA estimates that 85–90% of US GHG emissions are covered under the GHGRP. Other recent studies, however, argue that the GHGRP is less complete than estimated by EPA for two reasons (e.g., Kort et al., 2014; Karion et al., 2015; Lan et al., 2015; Lavoie et al., 2015; Lyon et al., 2015; Mitchell et al., 2015; Subramanian et al., 2015; Zimmerle et al., 2015). First, the emissions that are excluded from the GHGRP

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are sometimes larger than estimated by EPA, and second, the EFs used in the GHGRP are smaller than actual emissions from some source sectors like oil and natural gas.

2.3 Recent, direct measurements that support bottom-up efforts

Inventory development requires two different types of data: activity data and data that can be used to develop EFs. Activity data can come from economic, census, and remote sensing datasets, among other possible data sources. These datasets differ from those used to develop EFs. The IPCC provides a database of EF estimates but encourages countries to take measurements of emitters or emitting processes to develop tailored, country-specific EFs (Goodwin et al., 2006). A number of observation strategies can directly support the development and evaluation of country-specific EFs. We discuss a number of recent efforts here as well as the advantages and challenges of using these datasets.

One observation strategy is to measure GHG concentrations near an emitter or a group of emitters. These observations, by factor of their targeted spatial scale, can be directly used to evaluate a single source type and develop corresponding EFs. For example, a number of studies report on direct GHG measurements from individual facilities. These include direct stack measurements of power plant CO_2 emissions (e.g., Teichert et al., 2003) and numerous recent studies of CH_4 emissions from oil and gas operations: measurements of emissions from pneumatic controllers (Allen et al., 2015), compressor stations (Subramanian et al., 2015), transmissions and storage systems (Zimmerle et al., 2015), and abandoned wells (Kang et al., 2014). In addition, several site-level studies target agricultural emissions. Kebreab et al. (2008) and Sejian et al. (2010) review several measurement strategies, and Owen and Silver (2015) specifically review field studies on CH_4 emissions from manure.

On-road measurements provide a picture of emissions that is one spatial scale larger than direct facility observations. Existing studies often target oil and gas facilities (e.g., Roscioli et al., 2015; Brantley et al., 2014; Jackson et al., 2014; Lan et al., 2015; Mitchell et al., 2015; Subramanian et al., 2015) and mobile CO_2 emissions (e.g., Brondfield et al., 2012; Maness et al., 2015). In the case of oil and gas emissions, Brantley et al. (2014) explain that mobile measurements capture an integrated plume that includes all leaks from a given facility but rarely indicate which components caused those leaks.

The use of facility-level and on-road observations entails a number of challenges. For example, facility-level observations provide the most insight into detailed emissions processes from specific source sectors but can miss emissions events or processes. Observations of oil and gas facilities provide a prime example; scientists may not know about some leaks and therefore may not measure them, other leaks may be in inaccessible locations (e.g., Subramanian et al., 2015), and the largest leaks often come from ephemeral equipment failures at a small number of facilities that are difficult to identify (e.g., Brantley et al., 2014; Allen, 2014; Allen et al., 2015). Cost also limits direct measurements. For example, direct measurements from smokestacks are expensive, are typically only used for large point sources, and are generally not used in existing inventory estimates (National Research Council, 2010).

These observation strategies also require extrapolation to produce state or national-scale EF estimates. The relationship between activity data and emissions can be complex or spatially variable, making it difficult to extrapolate facility or on-road measurements. For example, CH_4 emissions from oil and gas are likely dominated by a small number of malfunctioning facilities. As a result, it is difficult to develop robust, national-scale EFs from a modestly-sized sample of facilities (Allen,

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2014). Furthermore, Brantley et al. (2014) explain that these leaks do not correlate with production and can vary greatly in time. Different oil and gas drilling basins also have different overall leak rates – from 0.3% in Pennsylvania's Mercellus shale region to 8.9% in Utah's Uintah basin (e.g., Karion et al., 2013; Petron et al., 2014; Karion et al., 2015; Peischl et al., 2015). These factors make it challenging to create consistent, generalizable EFs that can translate activity data into emissions.

These considerations also apply to other source sectors beyond the oil and gas industries. For example, grazing and manure management practices differ by region, and manure and landfill CH_4 emissions also differ by climate (US EPA, 2016a, ch. 5), all of which make extrapolation more challenging.

2.4 Impact of recent advances

Inventory estimates of sector-specific CO_2 emissions from the US are likely relatively accurate at national-scale but have substantial uncertainties at the local and state levels. Ackerman and Sundquist (2008), for example, compared smokestack versus fuel-based CO_2 estimates for US power plants and found a mean absolute difference of 16.6% but only a 1.4% total difference at national scale. Furthermore, Gately et al. (2015) found biases of 100% or more at the urban scale in CO_2 emissions estimates for mobile sources. However, they estimated a US national total that was broadly consistent with other inventories like VULCAN.

By contrast, sector-specific CH_4 emissions are more challenging to estimate and existing inventories for the US are highly uncertain at state and national scales. For example, several top-down studies indicate that the California state inventory is likely too low by a factor of 1.3 to 1.9 (Jeong et al., 2013; Wecht et al., 2014b), and several top-down studies estimate emissions for oil and gas drilling regions of Utah and Colorado that are up to three times bottom-up estimates (e.g. Karion et al., 2013; Petron et al., 2014). Overall, total US CH_4 emissions are likely $\sim$50% larger than estimated by EDGAR or US EPA (Miller et al., 2013; Wecht et al., 2014a; Turner et al., 2015). Fig. 1 compares several inventory estimates of sector-specific CO_2 and CH_4 emissions. Existing CO_2 inventory estimates are broadly consistent while CH_4 estimates vary between inventories and among inventory versions.

CH_4 inventories are so uncertain, in part, because of the complexity of many anthropogenic CH_4 source sectors. For example, emissions factors for oil and gas operations are difficult to estimate because a small number of emitters often account for a large fraction of emissions (e.g., Allen, 2014; Brantley et al., 2014; Allen et al., 2015; Lan et al., 2015; Mitchell et al., 2015) and because there are so many points along the natural gas production, processing, transmission, and distribution cycle that leak methane (e.g., Kang et al., 2014; Allen et al., 2015; McKain et al., 2015; Subramanian et al., 2015; Zimmerle et al., 2015).

Much of the uncertainty in CH_4 inventories stems from difficulties developing accurate EFs. Brandt et al. (2014) writes, "... measurements for generating emission factors are expensive, which limits sample sizes and representativeness. Many EPA EFs have wide confidence intervals. And there are reasons to suspect sampling bias in EFs, as sampling has occurred at self-selected cooperating facilities." For example, EPA's EFs for natural gas pipelines are based on a limited number of samples from a 1996 EPA and Gas Research Institute study; these EFs have a confidence interval of $\pm$65% (Beusse et al., 2014). Beyond the oil and gas industry, Owen and Silver (2015) also argue that EFs for agriculture are insufficient. These estimates are based upon a small number of pilot or lab experiments that were not explicitly designed for GHG inventory development.

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3 Top-down, inverse modeling strategies

In this section, we discuss inverse modeling strategies – strategies that leverage observations of atmospheric GHG concentrations to infer emissions at the Earth's surface. We specifically focus on strategies that attempt to parse the contribution of specific source sectors. The first part of this discussion (Sects. 3.1 – 3.2) focuses on efforts at local, urban, and regional scales. These studies do not provide direct state- or national-level estimates but could be combined or extrapolated to quantify emissions at larger spatial scales. Many studies in this category target source sectors that do not overlap spatially, at least at the spatial scale of interest. The second part of this discussion (Sects. 3.3 – 3.4) explores inverse modeling efforts that directly estimate sector-specific emissions at the state and national level. These efforts use observation networks that are sensitive to emissions across broad geographic regions, but these efforts must also devise strategies to disentangle emissions from multiple, spatially overlapping source sectors.

3.1 Local-scale inverse modeling

Local-scale inverse modeling can best attribute emissions when the study region has a single, dominant source type. An estimate of total emissions for the region thus provides insight into the source sector of interest.

Studies that fall within this category often employ one of a few different strategies to estimate emissions. For example, many efforts use a simple box-modeling approach to estimate emissions (e.g., Turnbull et al., 2011; Karion et al., 2013; Caulton et al., 2014; Karion et al., 2015; Schneising et al., 2014; Cambaliza et al., 2015; Peischl et al., 2015) while others use an atmospheric transport model to relate GHG observations to emissions (e.g., McKain et al., 2012, 2015). Studies that use the former strategy typically estimate emissions in a few steps: first, make GHG measurements upwind and downwind of the region of interest. Second, use the difference between these measurements, the rate of flow through the "box" (i.e., wind speed adjusted by pressure), and the volume of the box (i.e., the area of the box and the mixing height of the atmosphere) to calculate total emissions in the box. Most studies that use box modeling estimate a total flux for the region of interest, a number that is not spatially resolved.

Other studies in this category use a more involved approach: model atmospheric GHG concentrations using an emissions inventory and an atmospheric transport model. Subsequently, one can scale the inventory such that modeled concentrations reproduce measured atmospheric concentrations:

$$\mathbf{y} = H(\mathbf{x}^a) + \epsilon \quad (1)$$

$$\mathbf{x}^a = \beta \mathbf{x}^b \quad (2)$$

In these equations, $\mathbf{y}$ is an $n \times 1$ vector of atmospheric GHG observations. The function $H()$ is an atmospheric transport model that relates the surface emissions ($\mathbf{x}$), $((m_s \times m_t) \times 1)$ to the observations ($\mathbf{y}$). The variable m_s denotes the number of model grid boxes in space, and m_t denotes the number of time periods. In one study, this emissions estimate varied both spatial and temporally (McKain et al., 2012), and in another study, the emissions varied spatially but were constant in time ($m_t = 1$)

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(McKain et al., 2015). The superscripts a and b denote an emissions inventory and final emissions estimate, respectively. In addition, ϵ is an $n \times 1$ vector of errors (e.g., errors in estimated transport, in the measurements, and in the estimated emissions, among other errors). The objective of this approach is to scale an inventory estimate ($\mathbf{x}^b$, using a single scaling factor β) so that the modeled GHG concentrations ($H(\mathbf{x}^a)$) reproduce observed concentrations ($\mathbf{y}$).

These local-scale efforts can target sources with very large emissions or very uncertain emissions. For example, numerous existing studies have targeted emissions from cities. Cities account for 70% of global fossil fuel CO_2 emissions, so insight into urban emissions provides insight into a large fraction of total anthropogenic GHG emissions (US Energy Information Administration (EIA), 2016). Note that studies in this category generally do not discriminate among different urban source sectors but can provide insight into the contribution of urban CO_2 sources versus power plant CO_2 sources (which often occur well outside city limits). Existing efforts have estimated CO_2 emissions for Indianapolis, Indiana (Mays et al., 2009); Sacramento, California (Turnbull et al., 2011); and Salt Lake City, Utah (McKain et al., 2012) as well as CH_4 emissions from Boston, Massachusetts (McKain et al., 2015) and Indianapolis (Cambaliza et al., 2015). McKain et al. (2012) and McKain et al. (2015) used the approach in Eq. 1 while the other studies implemented box models.

Other studies in this category target oil and natural gas industry emissions. Existing studies have used aircraft observations to estimate CH_4 emissions from Utah's Uintah drilling basin (Karion et al., 2013), from southwest Pennsylvania (Caulton et al., 2014), from Colorado's Denver-Julesburg Basin (Petron et al., 2014), from the Barnett Shale in Texas (Karion et al., 2015; Lavoie et al., 2015), and from the Haynesville, Fayetteville, and Marcellus shale regions (in Texas, Arkansas, and Pennsylvania, respectively) (Peischl et al., 2015). In addition to these aircraft-based studies, one study used the SCIAMACHY satellite to estimate CH_4 emissions from the Eagle Ford and Bakken shale regions in Texas and North Dakota, respectively (Schneising et al., 2014). Several of these studies found leak rates that greatly exceed EPA's estimated emissions factors (e.g., Karion et al., 2013; Petron et al., 2014; Schneising et al., 2014) while other studies estimate leak rates that are comparable to EPA's numbers (e.g., Caulton et al., 2014; Peischl et al., 2015). Differences in drilling technology and practices from one basin to another may account for these contrasting results (e.g., Peischl et al., 2015).

These local-scale inverse modeling studies confer a number of advantages relative to other top-down strategies. These strategies capture emissions from all facilities in a given region, including those with anomalously high emissions. In the past, EPA has had difficulty designing facility-level measurements that adequately sample these anomalous emitters (Sect. 2.4). An additional advantage of these strategies is their ease of implementation relative to those discussed in subsequent sections (Sects. 3.3 – 3.4). Box modeling requires an estimate of air flow into and out of the box, but this approach does not require a full atmospheric transport model. Furthermore, the strategies discussed in this section are not as computationally intensive as many of the state- and national-scale strategies discussed later in Sect. 3.3.

These strategies also bring a number of challenges. A locality or region must have a single, dominant source sector or have spatially (or temporally) non-overlapping source sectors in order to attribute emissions using this strategy (e.g., Hutyra et al., 2014; Peischl et al., 2015). For example, Peischl et al. (2015) estimated oil and gas emissions from drilling regions that also contain livestock, landfills, and wastewater treatment facilities, all of which produce CH_4 emissions. The authors subtracted an inventory estimate of these non-hydrocarbon CH_4 sources from their estimated emissions total, and they attributed

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the remaining emissions to oil and gas activities. The authors point out that these non oil and gas source sectors are small contributors relative to oil and gas operations (8.5 – 19% of the CH_4 emissions total in each region), and uncertainties in these other source sectors would likely have a small impact on their oil and gas emissions estimate.

Complex environmental conditions can also pose a challenge for local-scale inverse modelings strategies, particularly for box models. A simple box modeling setup can be difficult to apply when atmospheric advection, vertical mixing, or upwind "clean air" measurements are highly heterogeneous across the box; these quantities should not contain patterns that are difficult to capture using a small number of parameters. For example, Turnbull et al. (2011) report that their CO_2 budget for Sacramento, estimated using a box model, is uncertain by a factor of two due to uncertainties in estimated wind speed and upwind "clean air" concentrations. Furthermore, Karion et al. (2015) estimated CH_4 emissions for the Barnett Shale that varied from 4.4×10^4 to 10.9×10^4 kg hr^{-1}, depending on the flight. However, the authors explain that two of the eight flights occurred during non-ideal meteorological conditions, and the range of estimates collapses to 6.1×10^4 to 8.8×10^4 kg hr^{-1} when those flights are excluded from the analysis. Atmospheric transport models can simulate more complex atmospheric transport patterns but still have difficulty modeling local- or urban-scale phenomena, including small-scale turbulent eddies, air flow through street canyons, and vertical mixing in a human-built landscape (e.g. Nehrkorn et al., 2013). These modeling challenges also apply to the state- and national-scale strategies discussed in Sects. 3.3 – 3.4. New innovations in atmospheric monitoring and instrumentation may reduce some of these uncertainties. Cambaliza et al. (2014), for example, explain that LIDAR instruments can measure atmospheric mixing height, and LIDAR deployment could therefore improve certain aspects of atmospheric modeling, particularly at local and regional scales.

3.2 Observations that support local-scale inverse modeling

Many recent, local-scale observation efforts have focused on urban monitoring and on oil and gas basins. Existing urban, atmospheric measurement networks include Salt Lake City, Utah (McKain et al., 2012); Los Angeles, California (Duren, 2016); Oakland, California (Cohen, 2016), the Bay Area Air Quality Management District (Fairley and Fischer, 2015), and Indianapolis (Mays et al., 2009; Cambaliza et al., 2015). Recent local-scale aircraft campaigns include the INFLUX campaign focused on the Indianapolis metro region (Cambaliza et al., 2015), the SENEX and SOGNEX campaigns focused on multiple oil and gas drilling basins (Peischl et al., 2015; NOAA Chemical Sciences Division, 2016), and the Barnett Coordinated Campaign (Smith et al., 2015; Karion et al., 2015) (Fig. 2). In addition to these urban and oil and gas studies, Lindenmaier et al. (2014) used ground-based, CO_2 column observations to identify emissions from a large coal-fired power plant in the Four Corners region of the western US.

The observational strategies described above are relatively diverse. These efforts include a combination of aircraft and stationary sites (e.g., telecommunications towers or building rooftops). Some of these campaigns provide a one or two day snapshot in time (e.g, most oil and gas studies) while other campaigns involve sustained measurements over a year or more (e.g., urban observation networks like LA Megacities and the Indianapolis INFLUX project).

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3.3 State- and national-scale inverse modeling

The top-down strategies discussed in this section provide sector-specific GHG emissions estimates across larger regions, regions that typically have several overlapping source sectors. Furthermore, these strategies make updates to the emissions estimate that are spatially resolved in some way. The three strategies discussed in this section use both GHG observations and inventories to attribute sector-specific emissions. Each approach, however, use a different mix; the first approach relies most heavily on existing inventories while the last relies most on GHG observations.

One strategy used by several studies will scale the individual source sectors in a bottom-up inventory. This setup is often similar to a multiple regression:

$$\mathbf{x}^a = \sum_i^p \beta_i \mathbf{x}_i^b \tag{3}$$

where i denotes an individual source sector from a bottom-up inventory, and p indicates the total number of source sectors in the inverse model. The observational constraint ($\mathbf{y}$) in this approach is the same as in Eq. 1. This setup also assumes that each $\mathbf{x}_i^b$ ($(m_s \times m_t) \times 1$) is defined at all spatial locations and is defined for all time periods. In one study, each $\mathbf{x}_i^b$ was spatially but not temporally resolved (e.g., $m_t = 1$) (Zhao et al., 2009), while in another study, $\mathbf{x}_i^b$ was resolved in both space and time (Jeong et al., 2013). The unknown scaling factors (β_i) adjust the magnitude of different source sectors in the bottom-up inventory; these factors are estimated by the inverse model. As a result of this setup, the estimated emissions ($\mathbf{x}^a$) will always be a linear combination of source-specific emissions patterns in an existing bottom-up inventory. Studies that use this approach often estimate the scaling factors (β_i) using Bayesian statistics; these frameworks can weigh uncertainty in the measurements ($\mathbf{y}$) and in the atmospheric model ($H()$) against uncertainty in the initial or prior guess for the scaling factors (This guess is typically unity.) (e.g., Rayner et al., 2016).

To date, a handful of studies have leveraged this approach to attribute emissions of CH_4. For example, Zhao et al. (2009) and Jeong et al. (2013) used atmospheric measurements from tall towers to estimate emissions from individual source sectors in California. Both studies found higher CH_4 emissions from agriculture relative to the EDGAR emissions inventory.

This scaling factor approach brings several strengths and weaknesses. An advantage of this approach is that it not only provides an estimate of total emissions but also the contributions of individual source sectors. The approach can be relatively easy to implement from a statistical perspective. The statistics are similar to a multiple linear regression. With that said, one still needs to run an atmospheric transport model once per source sector to create $H()$ and must have an estimate of background or upwind, clean air concentrations.

A notable challenge of this strategy is that it requires accurate knowledge of the spatial distribution of each source sector. The estimated emissions will always be a linear combination of source-specific emissions patterns from an existing inventory, and errors in the spatial distribution of these inventories will propagate into errors in sector-specific attribution. Furthermore, the atmospheric GHG observations ($\mathbf{y}$) must be sensitive to differences in the space-time patterns among different source sectors. Worded differently, the column vectors $H(\mathbf{x}_i^b)$ must be distinct from one another, and each column must explain substantial variability in $\mathbf{y}$. If the former condition does not hold, then the individual source sectors $\mathbf{x}_i^b$ are collinear; colinearity can lead to

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unphysical scaling factors (β_i) and unrealistically large uncertainty estimates (e.g., Zucchini, 2000). If the latter condition does not hold, then the scaling factors may be poorly constrained by the data, resulting in uncertain or unrealistic sector-specific estimates. To account for these challenges, Jeong et al. (2013) only reported source-specific estimates when they obtained scaling factors that were statistically significantly different from zero.

A second common inverse modeling strategy will scale an emissions inventory at the model grid level to better reproduce the atmospheric observations ($\mathbf{y}$). All of the strategies discussed previously scale the spatial patterns in an existing inventory. By contrast, this strategy estimates an emissions level for each location in the model domain, and the resulting estimate can have spatial patterns that are different from any inventory. Existing studies in this category have constructed inversions in slightly different ways, but most have used bottom-up inventory estimates ($\mathbf{x}^b$, dimensions $(m_s \times m_t) \times 1$) that are spatially and temporally variable (e.g., Wecht et al., 2014a, b; Turner et al., 2015). The scaling factors (β) in these studies, by contrast, were spatially variable but temporally constant. These estimates have the following general form:

$$\mathbf{x}^a = (\mathbf{1}_{m_t} \otimes \beta)\mathbf{x}^b \tag{4}$$

where β is a $m_s \times 1$ vector of scaling factors, $\mathbf{1}_{m_t}$ is a $m_t \times 1$ vector of ones, and $\otimes$ is a Kronecker product that repeats the vector of scaling factors (β) for each of m_t time periods. The observational constraint ($\mathbf{y}$) in this approach is the same as in Eq. 1. This approach is also Bayesian in nature; the modeler sets an initial guess for the scaling factors (typically unity) and an uncertainty in that initial guess; this information guides the estimate for β, particularly when the scaling factors (β) are under-constrained by the available observations ($\mathbf{y}$) (e.g., Rayner et al., 2016).

This approach does not support source attribution in and of itself; the initial guess ($\mathbf{x}^b$) and the scaling factors (β) are broken down by location but not by source sector (though the inventory underlying $\mathbf{x}^b$ may provide sector-specific information). However, several studies have adapted this strategy to support sector-specific attribution. These studies attribute the emissions in $\mathbf{x}^a$ grid box by grid box using the relative magnitude of each emissions source in the bottom-up inventory:

$$\mathbf{x}_i^a = (\mathbf{1}_{m_t} \otimes \beta)\mathbf{x}_i^b \tag{5}$$

As a result of this setup, the relative magnitude of the source sectors in any one grid box will be the same as in the bottom-up inventory.

Wecht et al. (2014b) leveraged this strategy to estimate CH_4 emissions for California using measurements from the CALNEX aircraft campaign. Like Zhao et al. (2009) and Jeong et al. (2013), they also found higher emissions from agriculture relative to EDGAR. Wecht et al. (2014a) and Turner et al. (2015) further applied this strategy to attribute emissions at continental scales; these studies used the SCIAMACHY and GOSAT satellites, respectively, to estimate sector-specific CH_4 emissions across North America. Both studies estimated larger emissions from agriculture relative to the EPA and EDGAR inventories. Turner et al. (2015) estimated oil and gas emissions that are a factor of two larger than EDGAR while Wecht et al. (2014a) found that these emissions are broadly consistent with EDGAR.

This strategy has a number of advantages and weaknesses relative to other approaches. The strategy can be used to estimate emissions at grid scale, and the resulting emissions estimate will not be the a linear combination of existing inventory estimates.

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However, it assumes that the inventory has correctly estimated the relative magnitude of each emissions source in each model grid box. Errors in this relative magnitude will produce errors in the sector-specific attribution.

Third, and finally, a number of studies have leveraged a strategy known as geostatistical inverse modeling (GIM) to estimate GHG fluxes generally (e.g., Michalak et al., 2004; Gourdji et al., 2008, 2012) and anthropogenic emissions specifically (Miller et al., 2013, 2016; Shiga et al., 2014; ASCENDS Ad Hoc Science Definition Team, 2015). This approach will attribute patterns in the emissions to individual anthropogenic source sectors when possible. However, it will leave emissions as unattributable when those emissions do not match the space-time patterns in any bottom-up inventory or when the information content of the atmospheric observations is insufficient for attribution:

$$\mathbf{x}^a = \sum_i^p \beta_i \mathbf{x}_i^b + \xi \tag{6}$$

The vectors $\mathbf{x}_i^b$ can be individual source sectors from a bottom-up inventory (similar to Eq. 3). The inverse model will then map the emissions on to those patterns to the extent possible. Additionally, patterns in the atmospheric observations ($\mathbf{y}$) may not always match patterns in an existing inventory ($H(\mathbf{x}_i^b)$). The inverse model will further add (or subtract) emissions at the model grid scale to better reproduce the atmospheric observations ($\mathbf{y}$). These emissions are denoted by the vector ξ ($(m_s \times m_t) \times 1$), and a GIM typically labels the emissions in ξ as unattributable. Furthermore, existing studies allow $\mathbf{x}_i^b$ and ξ to vary both spatially and temporally (Miller et al., 2013; Shiga et al., 2014; ASCENDS Ad Hoc Science Definition Team, 2015), in contrast to the studies described earlier in this section.

Several studies have leveraged this strategy in the context of both anthropogenic CH_4 and CO_2 emissions. Miller et al. (2013) used a GIM and in situ atmospheric measurements to estimate sector-specific CH_4 emissions in the US; like Turner et al. (2015), they found higher emissions from the agriculture and oil and gas sectors relative to inventory estimates. Miller et al. (2016) also used this strategy to separate CH_4 emissions patterns due to wetlands from anthropogenic emissions and to evaluate bottom-up estimates of the former emissions category. Two studies (Shiga et al., 2014; ASCENDS Ad Hoc Science Definition Team, 2015) implemented a GIM-based framework to identify anthropogenic CO_2 emission patterns using in situ and satellite CO_2 observations, respectively. They investigated whether the atmospheric signal resulting from anthropogenic CO_2 emissions could be reliably identified given the confounding signal from biospheric CO_2 fluxes. They found that in situ and remote sensing CO_2 networks could only identify anthropogenic emissions in a few regions during a few months of the year. This identification was hampered by biospheric CO_2 fluxes, by atmospheric transport errors, and by the sparsity or quality of the CO_2 observations.

The GIM approach makes more conservative assumptions relative to other source attribution strategies discussed in this section. A GIM will only attribute emissions to patterns in a bottom-up inventory when that inventory matches patterns in the atmospheric GHG observations. In Miller et al. (2013), for example, the GIM mapped 60% of total US CH_4 emissions onto patterns in the EDGAR inventory and found that 40% of the total emissions were unattributable to the patterns in any bottom-up dataset. By contrast, the other approaches discussed above will attribute 100% of the emissions. In GIM studies like Miller et al. (2013), the unattributable emissions indicate shortfalls in either the greenhouse gas observation network or available bottom-up data. In the former case, existing atmospheric observations do not provide enough information to

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reliably estimate sector-specific emissions patterns. For example, the information content of the atmospheric observations in Miller et al. (2013) was insufficient to uniquely constrain emissions from coal mining, and those emissions were included in ξ instead of $\sum_i^p \beta_i \mathbf{x}_i^b$. In the latter case, the unattributable emissions in ξ indicate inaccuracies in the spatial distribution of available inventory estimates. Existing inventories do not have well-developed activity data for the oil and gas industry, and the unattributable emissions in Miller et al. (2013) provide information about shortfalls in these activity datasets.

Overall, existing regional- to national-scale studies have been far more successful at attribution for CH_4 than CO_2, irrespective of the inverse modeling strategy. Biospheric CO_2 fluxes are large relative to anthropogenic CO_2 emissions at diel to monthly time scales, particularly during the growing season, and the spatial and temporal distribution of these fluxes is highly uncertain (e.g., Huntzinger et al., 2012). The inverse modeling strategies in this section would therefore be difficult to apply to CO_2, unless one chose an arid study region or estimated emissions in winter when biospheric fluxes are small. According to Shiga et al. (2014), the patterns in $\mathbf{x}_i^b$ corresponding to anthropogenic CO_2 emissions rarely explain substantial variability in atmospheric CO_2 observations.

3.4 Observations that have been used to attribute emissions at state and national scales

The observations discussed in this section do not provide a direct constraint on an individual source sector but have been used by existing regional- and national-scale inverse modeling studies (Sect. 3.3) to support sector-specific attribution. These observations are typically distributed across a broad geographic region. They are therefore sensitive to emissions over a large area and can constrain larger regions, albeit with less detail than the local approaches discussed in Sect. 3.2.

Observations in this category include air samples collected atop telecommunications towers and from aircraft: the NOAA tall tower observation network (Andrews et al., 2014), regular NOAA aircraft monitoring (Sweeney et al., 2015), the Environment and Climate Change Canada tower monitoring network (Environment and Climate Change Canada, 2011), the California Greenhouse Gas Research Monitoring Network (e.g., Zhao et al., 2009; Jeong et al., 2012, 2013), and a privately-funded tower network operated by Earth Networks (Fig. 2). Most of the inverse modeling studies discussed in the previous section (Sect. 3.3) used these in situ observation networks to estimate sector-specific emissions (Zhao et al., 2009; Jeong et al., 2013; Miller et al., 2013; Shiga et al., 2014; ASCENDS Ad Hoc Science Definition Team, 2015).

Several satellites make total column observations of GHG concentrations: observations of CO_2 (e.g., AIRS, SCIAMACHY, GOSAT, and OCO-2) and of CH_4 (e.g., SCIAMACHY, AIRS, TES, IASI, GOSAT) (Fig. 2). Streets et al. (2013) describe each of these satellites and the respective measurement characteristics in detail. Only a handful of studies have used these datasets to attribute sector-specific emissions in the US, and these existing studies focus on CH_4, not CO_2 (e.g., Wecht et al., 2014a, b; Turner et al., 2015). Furthermore, some remote sensing datasets are more sensitive to surface emissions and have smaller errors/biases relative to other datasets. TES and SCIAMACHY show limited ability to constrain surface emissions (Wecht et al., 2014a; Alexe et al., 2015). For example, Wecht et al. (2012) could not reproduce patterns in North American CH_4 emissions using synthetic, simulated observations from TES. GOSAT, by contrast, provides more promising results. Turner et al. (2015) used GOSAT observations to estimate sector-specific CH_4 emissions in North America and found results that were broadly consistent with emissions estimates derived from the US tall tower and aircraft monitoring network (Miller et al., 2013).

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4 Novel strategies that could be used for estimating sector-specific emissions

This section discusses two observational strategies to support top-down modeling efforts, strategies that show promise for estimating sector-specific emissions. First, we discuss the potential of upcoming and proposed satellite-based GHG observations. Next, we discuss the utility of 'secondary tracers.' These gases or isotopologues are co-emitted with GHGs and aid in sector-specific attribution.

4.1 New satellite-based GHG observations

An increasing number of satellites collect observations of total column CO_2 and CH_4, and several more missions are planned for future years (e.g., TROPOMI and ASCENDS). However, the potential of existing and upcoming space-based observations for constraining anthropogenic emissions is not yet clear.

Existing studies are mixed on whether current and proposed satellites can identify patterns from anthropogenic CO_2 emissions. These studies generally examine the detectability of total fossil fuel CO_2 emissions, a less ambitious goal than monitoring specific source sectors. Several provide a positive outlook. For example, Schneising et al. (2008) report a detectable, 1.5ppm CO_2 column measured by SCIAMACHY over an industrial region of Germany. Kort et al. (2012) and Schneising et al. (2013) argue that GOSAT and SCIAMACHY, respectively, can detect fossil fuel CO_2 emissions from large urban regions, using several global cities as case studies. In addition, the National Research Council (2010) predicts that the OCO-2 satellite will be sufficient to constrain emissions from very large coal power plants.

Other existing studies offer a more skeptical perspective on the utility of satellite-based CO_2 observations. This utility is limited by measurement noise, measurement biases, the spatial and temporal sparsity of observations, and the limited sensitivity of some observations to the near-surface atmosphere. Keppel-Aleks et al. (2013) argue that variations in total column CO_2 are largely obscured by biospheric fluxes and that remote sensing observations would therefore have limited ability to constrain fossil fuel emissions. Furthermore, Gavrilov and Timofeev (2015) found large biases (4.7 ± 2.6 ppm) in GOSAT observations of CO_2 at a spectrometer site in Russia. Future improvements in retrieval algorithms, however, could decrease these biases.

Planned, future satellite observations may be even more capable at supporting efforts to estimate fossil fuel CO_2 and CH_4 emissions. Observations from ASCENDS, a future LIDAR-based satellite mission, would likely support evaluation of fossil fuel CO_2 emissions from the US East Coast (ASCENDS Ad Hoc Science Definition Team, 2015) and could detect large changes in emissions from broad regions like Europe or China (Hammerling et al., 2015). Furthermore, a proposed, future geostationary satellite mission could potentially constrain emissions from large urban regions like Shanghai, China (Rayner et al., 2014).

New remote sensing observations of CH_4 also show promise. The forthcoming TROPOMI satellite is a project of the European Space Agency and is currently scheduled for launch in late 2016 (Veefkind et al., 2012). Wecht et al. (2014a) argue that observations from TROPOMI may have the same ability to constrain California CH_4 emissions as the recent, intensive CALNEX aircraft campaign.

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4.2 Secondary tracers

Secondary tracers are co-emitted with GHGs and are often emitted from only a small number of source sectors. These tracers make it possible to isolate and factor out at least a portion of natural fluxes or factor out emissions from source sectors that are not of primary interest. The top-down approaches discussed previously either require a limited geographic scope or accurate activity data to effectively estimate sector-specific emissions. Secondary tracers could identify sector-specific emissions without these limitations (though secondary tracers present challenges of their own). Examples of secondary tracers include radiocarbon, ethane, $^{13}CO_2$, $^{13}CH_4$, and carbon monoxide (CO). We focus on radiocarbon and ethane because they hold particular promise.

4.2.1 Radiocarbon

Radiocarbon (^{14}C) is produced by cosmic rays in the upper atmosphere and has a lifetime of approximately 5,730 y before decaying back to ^{12}C (Bowman, 1990). More recently, nuclear bomb testing has elevated ^{14}C within the atmosphere. CO_2 fluxes from the biosphere will mirror the isotopic composition of the atmosphere at the time that carbon was incorporated into the plant. CO_2 emissions from fossil fuels, by contrast, contain no ^{14}C because fossil fuel reservoirs are far older than the decay lifetime of ^{14}C, and these reservoirs have not interacted with atmospheric carbon during the intervening time period.

Several exploratory studies used radiocarbon to separate the atmospheric CO_2 signal from biogenic versus anthropogenic emissions. One study used radiocarbon measurements from the US East Coast to estimate the relative contribution of fossil fuel versus biogenic emissions (Miller et al., 2012). Another study reported on radiocarbon measurements in California (Riley et al., 2008). Graven et al. (2011) and LaFranchi et al. (2013) used radiocarbon observations from an aircraft and a tall tower, respectively, to estimate the contribution of anthropogenic and biogenic CO_2 emissions in Colorado. Beyond these studies, radiocarbon measurements are not widely used in regional- or continental-scale inversions.

These measurements have not been widely used, in part, because only a handful of atmospheric monitoring sites in the US report radiocarbon concentrations. An expanded observation network shows enormous potential. A handful of tall tower monitoring sites in the US report radiocarbon and only two regular US aircraft monitoring sites do (Basu et al., 2016). The National Research Council (2010) recommended that the US invest $15–20 million annually to build 10 radiocarbon monitoring stations across the US, but that goal has not yet come to fruition. A recent paper by Basu et al. (2016) argued that this level of investment would allow scientists to constrain US fossil fuel CO_2 emissions to within 1% per year and to within 5% per month.

Despite this promise, the use of atmospheric radiocarbon measurements also presents several challenges. One primary challenge is accounting for the disequilibrium effect (Bowman, 1990). Atmospheric concentrations of ^{14}C have changed in the past 75 years due to nuclear bomb testing. CO_2 from decomposing organic matter (heterotrophic respiration) will reflect ^{14}C levels during the time that carbon was incorporated into plant tissue, not current atmospheric levels of ^{14}C. Furthermore, the lifetime of dissolved gases in the ocean is much longer than 75 years, so the isotopic signature of air-sea gas exchange will also lag the recent rise in atmospheric ^{14}C. One must account for this mismatch or 'disequilibrium' when using radiocarbon measurements

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to partition between fossil fuel CO_2 and biospheric CO_2; biospheric (and ocean) fluxes will not necessarily match current atmospheric ^{14}C levels but rather reflect the levels of a past date.

4.2.2 Ethane

Methane is the primary component of natural gas, but natural gas also contains small quantitates of other alkanes, including ethane. These trace constituents are collectively referred to as natural gas liquids. Enhancements in atmospheric ethane concentrations indicate leaks from natural gas and oil infrastructure because these operations are a primary source of ethane to the atmosphere (e.g., Rudolph, 1995). Other CH_4 emitters, including agriculture, landfills, and wetlands do not emit higher order alkanes in substantial amounts. For example, Peischl et al. (2013) estimated that natural gas leaks account for 90% of all ethane emissions in the Los Angeles metro region. If one has an estimate of ethane emissions and an estimate of the ethane content of natural gas, then one can estimate CH_4 emissions from oil and gas infrastructure. McKain et al. (2015), for example, measured CH_4 and ethane at several sites in Boston, and they used CH_4-ethane ratios reported from natural gas pipeline operators to estimate the portion of Boston's CH_4 emissions that are due to natural gas leaks. Several other studies have similarly used ethane measurements to explore oil and gas industry emissions from Los Angeles (Wennberg et al., 2012), Dallas, Texas (Yacovitch et al., 2014), the Barnett shale region (Smith et al., 2015; Townsend-Small et al., 2015), and from global oil and gas operations (e.g., Simpson et al., 2012; Schwietzke et al., 2014).

The use of ethane for CH_4 source attribution brings several challenges. Until recently, ethane has been difficult to measure in the atmosphere. However, Aerodyne, Inc. now markets and ethane analyzer (Yacovitch et al., 2014), and NOAA has developed a new instrument for its monitoring network that includes ethane in the analysis. In addition, the ethane content of natural gas can vary by region and will change if natural gas liquids are removed at processing facilities (Fig. 3). These variations complicate the task of inferring CH_4 emissions using ethane measurements. Smith et al. (2015), for example, found three distinct ethane signatures in different areas of the Barnett shale region. Townsend-Small et al. (2015) report that emissions operations in the Barnett ranged from 6% ethane at natural gas wells to 13% ethane at oil wells.

In summary, secondary tracers like ethane and radiocarbon allow scientists to leverage measurements networks with broad spatial coverage (like those in Sect. 3.4) to estimate specific source sectors. These measurements bypass, to some degree, the need to rely on the spatial and temporal patterns in an inventory for source attribution and the need to have accurate activity data to support inverse modeling. With that said, only some CO_2 and CH_4 source sectors have obvious secondary tracers, and the associated atmospheric observations are primarily collected by in situ networks, not by satellites. Furthermore, progress in this area has been limited because of measurement availability, but this limitation could change in the future with more funding (i.e., in the case of radiocarbon) or deployment of new instrument technology (i.e., in the case of ethane).

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5 Synthesis discussion

In this section, we synthesize progress to date on estimating sector-specific CO_2 and CH_4 emissions at state and national scale. We also discuss forward-looking opportunities to improve sector-specific GHG emissions estimates, with a particular focus on opportunities to integrate bottom-up and top-down strategies.

Recent innovations in both bottom-up and top-down efforts have advanced scientists' abilities to identify emissions from specific source sectors. Several efforts have produced high resolution, sector-specific inventory products that are based on more accurate, detailed activity data and EFs. These products have largely been driven by research in academia and by the Joint Research Centre in Europe, not by US state or national governments. New inverse modeling strategies can incorporate these inventory estimates in more rigorous ways that are not limited to the spatial patterns in the inventory. In addition, more extensive observations are available to support these inverse modeling efforts, observations that span a number of spatial scales. For example, numerous intensive measurement campaigns in the past five years have focused on large GHG-emitting regions, particularly cities and oil and gas production basins. The national US in situ network and remote sensing GHG observations have also expanded in the last decade, though the US in situ network expansion is smaller than the level required for robust evaluation of a wide array of GHG source sectors.

Despite these advances in bottom-up inventories, top-down strategies, and measurement density, the scientific community has only been able to use inverse modeling and atmospheric data to improve sector-specific emissions estimates in a relatively small number of cases. To date, the community has had far more success integrating top-down and bottom-up estimates for CH_4 than for CO_2; the atmospheric signal from biospheric CO_2 fluxes often obscures the signal from fossil fuel emissions, except in some urban environments. CH_4 emissions inventories are far more uncertain than CO_2 inventories, and the community has been able to use top-down inverse modeling to improve these inventories when they arguably stood to benefit most.

Specifically, the community has been most successful with top-down, sector-specific attribution in two types of scenarios: intensive measurement campaigns paired with local-scale inverse modeling and opportunistic cases. In the former case, the community has put substantial resources into intensive, local-scale measurement campaigns for a few specific source sectors. Measurements from each affected locality or region provide a puzzle piece, and the community has begun to assemble a cohesive, national-scale picture by amalgamating these individual pieces. The community has employed this strategy in the case of CH_4 emissions from oil and gas operations (e.g., the SENEX, SONGNEX, Barnett Coordinated Campaign, etc.) and, to a lesser degree, in the case of urban CO_2 emissions (including recent measurement efforts in Los Angeles, Salt Lake City, Boston, and Oakland).

Other cases of successful source attribution have been largely opportunistic. In certain cases, the community had the right atmospheric measurements and spatially-distinct source sectors to attribute emissions at large spatial scales. For example, Miller et al. (2013) found large CH_4 emissions in Texas and Oklahoma that did not fit the spatial distribution of cows, and CH_4 measurements in that region correlated with measurements of higher order alkanes. The authors concluded that a large fraction of those emissions were likely due to oil and gas operations. A more recent study using satellite observations from GOSAT reached similar conclusions (Turner et al., 2015).

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Numerous future opportunities would improve scientists' ability to merge bottom-up inventories, inverse modeling, and GHG concentration data for better GHG source attribution:

1. Combine the strengths of existing datasets

The majority of inverse modeling studies to date have used only in situ or satellite GHG data to estimate emissions. Methane inverse modeling studies for North America provide a good example. Miller et al. (2013) used in situ observations from long term monitoring stations, Wecht et al. (2014a) used remote sensing observations from SCIAMACHY, and Turner et al. (2015) used remote sensing observations from GOSAT. Future studies may be able to attribute emissions more effectively by leveraging the strengths of all available in situ and remote sensing datasets. Different datasets often bring complementary strengths for this attribution: remote sensing datasets have broad spatial coverage and in situ datasets have complete temporal coverage and greater sensitivity to surface emissions, among other strengths. A number of challenges may have prevented the synthesis of multiple datasets in past studies: large datasets entail a number of computational challenges, the data are not always accessible (e.g., data from Environment Canada are not publicly available), and the observations can have different information content or error characteristics that are challenging to balance in a single framework. Future efforts that can combine these disparate datasets likely stand the best chance of attributing emissions to specific source sectors.

2. Expand several existing measurement strategies

Expanded GHG measurements would also advance efforts to attribute emissions to specific source sectors. As discussed earlier, some of the most successful top-down efforts to attribute emissions have been intensive aircraft campaigns. These campaigns are more flexible than the long term monitoring network and can easily target source sectors of interest by flying in specific regions, in flight patterns that encapsulate the source of interest, and by flying at certain times of year that have fewer competing biogenic sources. An expansion of these campaigns would enable scientists to target specific source sectors, including CO_2 emissions from large power plants, CH_4 from agriculture, and CH_4 from coal mines, among other source sectors. These aircraft campaigns could then be used to estimate regional-scale EFs. Existing aircraft campaigns, for example, have have estimated CH_4 leak rates for a range of different oil and gas drilling basins (see Sects. 3.1 – 3.2). The long term in situ atmospheric network and GHG monitoring satellites could be used to intelligently extrapolate and gap-fill these regional EFs at larger spatial scales and to identify broad trends over time.

In addition, successful cases of sector-specific attribution have usually involved observations that span multiple spatial and temporal scales. This strategy allows scientists to bridge between the regional scale that atmospheric observations are best able to constrain and the facility-level scale where inventories are strongest. For example, atmospheric observations can be used to identify regional differences between top-down and bottom-up estimates. Subsequent facility-level and on-road measurements can indicate why those regional differences occurred and how to improve EFs in a way that will bring inventories into agreement with top-down estimates. This measurement strategy can be expensive and requires extensive coordination, but it has been used successfully in the case of oil and gas CH_4 emissions (e.g., Allen, 2014; Brandt et al., 2014; Peischl et al., 2015). Bottom-up and top-down estimates of these emissions disagree at regional and national spatial scales (e.g., Miller et al., 2013; Turner et al., 2015). Subsequent facility and on-road measurements revealed that a small number of facilities account for a large percentage

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of emissions; EFs that account for this skewed distribution are more consistent with regional top-down estimates (e.g., Brantley et al., 2014; Lavoie et al., 2015; Subramanian et al., 2015).

Effective source attribution will also likely require the use of secondary tracers. Measurements of some secondary tracers, like ethane, have expanded markedly in the past several years with advances in instrumentation. With that said, measurements of tracers like radiocarbon are only available for some of the long term US monitoring sites.

3. Improve inverse modeling strategies with an eye toward secondary tracers

The inverse modeling community has yet to develop inverse modeling strategies that can fully leverage observations of secondary tracers. This task is not straightforward and would likely require the development of new strategies. These strategies would need to quantify heterogeneities in the ethane content of natural gas or the disequilibrium effect in the case of radiocarbon. Furthermore, these strategies would need to relate the primary and secondary tracers in a single statistical framework and would need to account for uncertainties in that relationship. Observations of these secondary tracers have historically been very sparse, so few studies have focused on designing statistical inverse modeling frameworks to fully exploit these tracers.

4. Develop detailed activity data as part of bottom-up efforts

Top-down efforts, like those outlined above, can help in developing regional-scale EFs for different source sectors. These studies can be particularly helpful when EFs are challenging to determine at facility scale. For example, direct measurements of oil and gas facilities are difficult to design because a small number of leaks account for the majority of emissions, and these large emitters may be difficult to find and/or representatively sample (see Sect. 2.3).

In contrast to EFs, activity data can only come from bottom-up inventory efforts. In fact, top-down efforts depend upon reliable activity data for attributing emissions (Sects. 3.1 and 3.3). Efforts to improve these activity datasets would markedly improve source attribution. In many cases, these activity data exist but are not publicly available or are not available in gridded form. Gurney et al. (2007) cite local fuel sales or electric utility bills as examples. CH_4 emissions from oil and gas provide an additional example. Oil and gas wells generally report production figures to state regulatory agencies, but this reporting varies by state, does not have a consistent format, and can be difficult to find (e.g., http://pmc.ucsc.edu/~brodsky/wellindex.html). The inaccessibility of accurate activity data for oil and gas operations has been a barrier to source attribution in recent national-scale CH_4 inverse modeling studies (Miller et al., 2013; Turner et al., 2015). These activity data are key to connecting inverse modeling results with bottom-up estimates of specific source sectors. Future bottom-up efforts should particularly focus on the development and public release of gridded activity data.

In synthesis, future improvements in bottom-up inventories and top-down strategies would likely complement one another and translate into more reliable, sector-specific emissions estimates; scientists will likely need to combine both strategies to robustly estimate GHG emissions from individual sources. Improved activity data would lead to gridded inventory estimates with more accurate spatial and temporal patterns. Top-down frameworks could then harness these patterns, along with more extensive, future GHG observations, to estimate regional-scale EFs for specific source sectors. National-scale observations of secondary tracers like radiocarbon and ethane would further strengthen these top-down efforts for applicable source sectors. This coordinated, combined approach offers the most promising opportunity to evaluate state and national GHG emissions reduction policies in the US.

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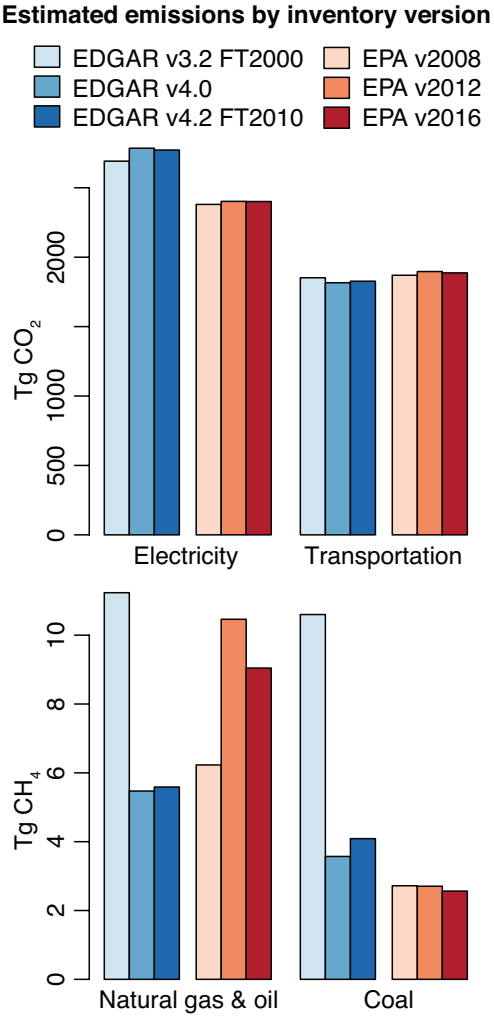

Figure 1. This figure displays several inventory emissions estimates for different US fossil fuel source sectors (Olivier et al., 2014; US EPA, 2016a). The figure includes both the EDGAR and EPA inventories as well as several versions of each inventory. All of the estimates are for 2005 except for EDGAR FT2000 which is for 2000. CO_2 emissions estimates are consistent from one inventory version to another and between EPA and EDGAR. Note that EDGAR includes CO_2 from heating in its electricity estimate while EPA does not. As a result, the EDGAR CO_2 estimate is higher than EPA's estimate. CH_4 estimates, however, vary widely between EPA and EDGAR and among inventory versions. These variations indicate how uncertain CH_4 inventory estimates are relative to CO_2 estimates.

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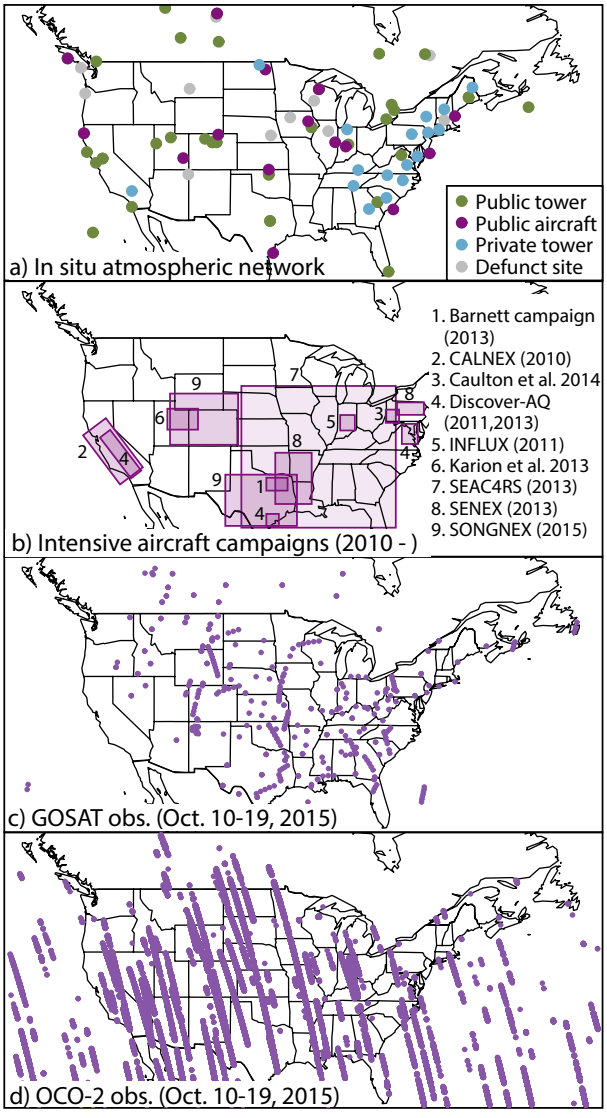

Figure 2. This figure displays existing CO_2 observations available from tower and regular aircraft sampling sites (**a**); from several recent, intensive aircraft campaigns (**b**); from the GOSAT satellite (**c**); and from the OCO-2 satellite (**d**). Note that the dots on each panel are not equivalent; an in situ monitoring site in panel **a** provides far more information than an individual CO_2 total column observation from GOSAT or OCO-2 (panels **c** and **d**, respectively). Public towers and public aircraft sites are operated by NOAA, DOE, Environment Canada, and partners. Private towers are operated by Earth Networks. Most tower and aircraft sites also include CH_4 observations.

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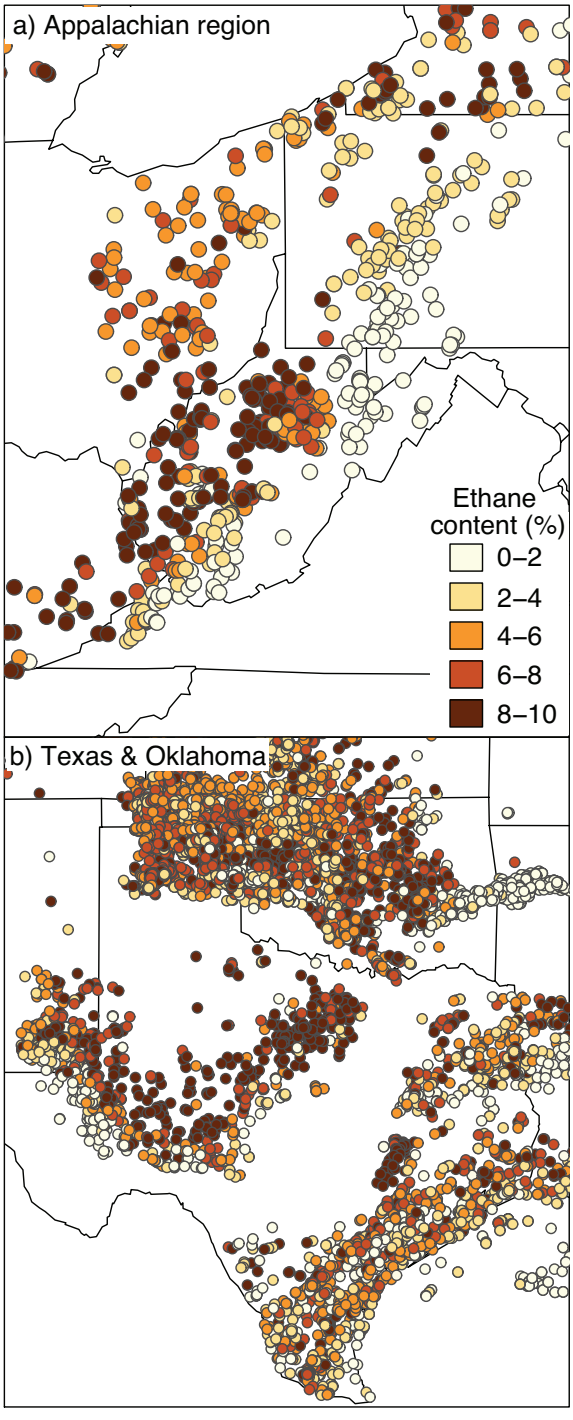

Figure 3. Ethane content of natural gas samples from the USGS Geochemistry Laboratory Database (USGS Energy Resources Program, 2015). Ethane content is a key parameter when estimating oil and gas CH_4 emissions using atmospheric ethane measurements. The samples show substantial heterogeneity in some regions (e.g., Oklahoma) and exhibit clear spatial patterns in other regions (e.g., Texas and West Virginia).