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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraining sector-specific CO_2 and CH_4 emissions in the United States" *by* Scot M. Miller and Anna M. Michalak

Scot M. Miller and Anna M. Michalak

scot.m.miller@gmail.com

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Thank you for the ideas and suggestions for the review paper. They have greatly helped us improve the manuscript. Below, we have listed each of the suggestions and the corresponding revisions that we have made to the manuscript.

- In the introduction there should be some mentioning of INDCs (Intended Nationally Determined Contributions), which were decided during COP 21 in Paris 2015.

  This is a great suggestion. We have included INDCs in the revised introduction.

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- P3 L1-2: I suggest reformulating to “frameworks that can synergistically leverage the information content of bottom-up datasets and top-down strategies using atmospheric GHG data”

  We have updated this sentence accordingly.

- P3 L7: May be reformulate “to attribute that trend to a specific source sector(s)” to e.g. “to attribute this trend to trends in specific source sectors”

  We have revised the sentence accordingly. The new wording sounds more precise.

- P4 L27: A reference for EDGAR needs to be included here.

  We have added a reference to EDGAR in this line.

- P15 L22: I think a reference to Dils et al., 2014, which systematically validates CH4 and CO2 products from GOSAT against TCCON data, would be appropriate: Dils, B., Buchwitz, M., Reuter, M., Schneising, O., Boesch, H., Parker, R., Guerlet, S., Aben, I., Blumenstock, T., Burrows, J. P., Butz, A., Deutscher, N. M., Frankenberg, C., Hase, F., Hasekamp, O. P., Heymann, J., De Mazière, M., Notholt, J., Sussmann, R., Warneke, T., Griffith, D., Sherlock, V. and Wunch, D.: The Greenhouse Gas Climate Change Initiative (GHG-CCI): comparative validation of GHG-CCI SCIAMACHY/ENVISAT and TANSO-FTS/GOSAT CO2 and CH4 retrieval algorithm products with measurements from the TCCON, Atmos. Meas. Tech., 7(6), 1723–1744, doi:10.5194/amt-7-1723- 2014, 2014.

  This is a great suggestion. We have added this reference to the corresponding line of the revised manuscript.

- P15 L28: Here I think the CarbonSat mission should be mentioned, as it combines high spatial resolution with a large swath, making it useful for emission detection. Some rel- evant papers are listed here: Buchwitz, M., Reuter, M.,

Bovensmann, H., Pillai, D., Hey- mann, J., Schneising, O., Rozanov, V., Krings, T., Burrows, J. P., Boesch, H., Gerbig, C., Meijer, Y. and Löscher, A.: Carbon Monitoring Satellite (CarbonSat): assessment of atmospheric CO2 and CH4 retrieval errors by error parameterization, Atmos. Meas. Tech., 6(12), 3477–3500, doi:10.5194/amt-6-3477-2013, 2013. Pillai, D., Buchwitz, M., Gerbig, C. and Koch, T.: Tracking city CO2 emissions from space using a high reso- lution inverse modeling approach: A case study for Berlin, Germany, Atmos. Chem. Phys., doi:10.5194/acp-16-9591-2016, 2016.

CarbonSat was a notable shortfall in the initial manuscript. We have added several lines on CarbonSat and GeoCARB to this section (along with the references above). Thank you for including these suggested references; they are very helpful.

- P17 L17: reword “now markets and ethane analyzer” -> “now markets an ethane analyzer”

  Thank you for pointing out this typo. We have fixed it in the revised manuscript.

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Thank you for the thorough and highly constructive suggestions on the manuscript. The suggestions are insightful and thoughtful and have been incredibly helpful for improving the manuscript.

- The scope of the review should be stated in the introduction. For example, many studies aimed at understanding CO2 uptake by terrestrial vegetation are evidently out of scope, even though biological CO2 sequestration may significantly offset US CO2 emissions.

  This is a great suggestion for clarifying the manuscript framing. We have added

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content to the introduction defining the scope as suggested here. The reviewer makes a great point about biological CO_2 sequestration. We felt that this topic would have expanded the scope of the review beyond what we could feasibly cover in a single paper. It would be an excellent topic for a future review paper, though.

- Discussion of the quality of satellite data required for anthropogenic flux estimation and trend estimation would be helpful. The measurement requirements to detect anthropogenic CO2 plumes are described in the 2010 NRC Report, Verifying Greenhouse Gas Emissions, and in publications describing the notional CarbonSat mission as well as in the CarbonSat report (http://esamultimedia.esa.int/docs/EarthObservation/SP1330-1_CarbonSat.pdf).

  We have added this information to Sect. 4.1 of the revised manuscript. In addition, we have also added mention of the newly announced GeoCARB satellite in this section.

- Also it should be noted that the current generation of satellite sensors are not designed to provide comprehensive global mapping and are therefore not ideally suited for urban and point/source estimation. OCO-2 and GOSAT were designed for global carbon cycle science rather than emissions monitoring. How does the uncertainty in e.g. the Kort et al. analysis of Los Angeles emissions using GOSAT compare with the requirements for useful urban trend detection (e.g., something like a 10

  The reviewer makes a great point, and we have added this information to Sect. 4.1. Current satellite data products are unlikely to detect a 10% trend over 10 years; the measurement uncertainties and retrieval biases associated with these products are likely too large relative to the XCO_2 increment. Kort et al. (2012) estimate that GOSAT could detect a trend as small as 22% from Los Angeles. However, Los Angeles is arguably an ideal case study, a very large city with a

small biospheric CO_2 signal.

- It would be useful to see more discussion about where existing inventories and/or inversions agree and where they disagree. For example, how do the Schneising et al SCIAMACHY fugitive methane emissions estimates for North America compare with those from aircraft campaigns?

  We have added text to Sect. 2.4 that highlights where existing estimates agree and disagree. Different top-down and inventory studies often have very different spatial scales and cover different time windows. These differences in scale can make disagreements among the estimates more challenging to identify. We also highlight this point in the revised version of Sect. 2.4.

- Finally, some more discussion of transport modeling errors would be useful. To what extent do uncertainties in simulated transport limit top-down flux estimation? What type of work is needed to address transport uncertainty?

  We have added content to the synthesis discussion in Sect. 5. We mention the importance of reducing transport errors and recent innovations that could aid in this effort (e.g., monitoring mixed layer height with LIDAR). In sections 3.1 and 3.3, we also highlight studies that discuss the impact of transport errors on sector-specific attribution. These studies include papers by Shiga et al. (2014) and Karion et al. (2015).

- page 2, line 20: Are there any regulations targeting CH4 emissions from agriculture? Perhaps worth mentioning here that agriculture is a large source of CH4 even if not regulated yet.

  To our knowledge, there are no regulations that mandate CH_4 emissions reductions from agriculture in the U.S.. In August of 2014, the US EPA, USDA, and US DOE released the "Biogas Opportunities Roadmap" targeting voluntary reduction strategies for agriculture

(https://www3.epa.gov/climatechange/Downloads/Biogas-Roadmap.pdf). We have updated this line of the manuscript with a brief mention of the roadmap.

- page 3, line 9: “meteorically” sounds sensational

  We have replaced this phrase with “began in the past decade.”

- page 3, line 15: Perhaps briefly discuss biological CO2 sinks and potential for deliberate sequestration, along with concomitant need for verification of such reservoirs. Also could mention challenges of accounting for emissions from CH4 wetlands, as well as CH4 emissions related to anthropogenic interference in the hydrological systems (emissions from reservoirs). Something about co-location of cows and oil and gas perhaps also worth mentioning here.

  We have added a sentence to this paragraph mentioning the possibility of biological or geological sequestration as a public policy tool and the need to verify those carbon sinks.

- page 4, line 1: I don’t see a reference for EDGAR inventory in this list of references for global efforts, though it is frequently used.

  We have added the following reference to EDGAR: European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL). Emission Database for Global Atmospheric Research (EDGAR), release version 4.3.1 http://edgar.jrc.ec.europa.eu/overview.php?v=431, 2016.

  This particular publication is one of the preferred references for EDGAR stated on their web site (http://edgar.jrc.ec.europa.eu/terms_of_use.php).

- page 4, line 10: For the example of coal gasification, how is energy lost in conversion of coal to gas taken into account? It seems like this should count as emissions from coal.

  We have clarified this line in the manuscript. This line does not refer to energy lost in the conversion of coal to gas. Rather, the energy converted from coal to

gas is moved from the “industrial other coal” category in EPA’s accounting to the “natural gas combustion” category. EPA explains, “The energy in this synthetic natural gas enters the natural gas distribution stream, and is accounted for in EIA natural gas combustion statistics. Because this energy of the synthetic natural gas is already accounted for as natural gas combustion, this amount of energy is deducted from the industrial coal consumption statistics to avoid double counting” (EPA 2016c, p. A-31).

- page 4, line 28: First mention of EDGAR, but I don’t see any reference. Perhaps add a url.

  We have added a reference to EDGAR (JRC/PBL 2016, as shown above).

- page 5, first paragraph: Perhaps mention for which years these products are available and how often they are updated (or not updated).

  We have added this information to the paragraph.

- page 5 line 8: instead of “rigorous” consider “detailed”

  We have replaced the words as suggested.

- page 5 line 14: repeated use of “EFs” results in confusing long sentence. Consider simplifying e.g., “...much higher EFs that result in higher emissions that are much more consistent. . .”

  We have simplified and shortened this sentence accordingly.

- page 5, various lines: over-use of the word “leverage” in this section

  We have reduced the usage of this word throughout this section.

- page 5, line 26: The Andres et al. effort is also government-sponsored

This statement is technically true since Andres works at Oak Ridge National Laboratory. What we intended to say is that most of these inventories are not constructed by regulatory agencies and are not part of an official regulatory agency inventory product. We have revised this line accordingly.

- page 5, line 31: “these omissions” since threshold plus ag exemption

  We have changed “this” to “these” as suggested.

- page 6, line 20: it would be helpful to define what is meant by on-road measurements, i.e. are these all ground-based mobile using public (or private) roads?

  All of the studies listed here use ground-based mobile measurements on roadways. Not all of these studies list whether the roads were public or private (e.g., Mitchell et al., 2015, Subramanian et al., 2015). With that said, many of the studies listed in the manuscript use public roadways (e.g., Brondfield et al., 2012; Brantley et al., 2014; Jackson et al., 2014; Lan et al., 2015; Maness et al., 2015; and Roscioli et al., 2015). Roscioli et al., (2015) also took ground-based mobile measurements within the perimeter of many sites.

- page 6, line 25: Smokestack measurements of CO2 are not used in the EPA inventory?

  We have edited this passage to make it more precise. EPA uses smokestack measurements in some contexts. For example, smokestack or facility level measurements are used in EPA’s Greenhouse Gas Reporting Program (GHGRP) (e.g., see Sect. 2.1 in https://www.epa.gov/sites/production/files/2016-03/documents/stationaryemissions_3_2016.pdf).

- page 7, line 2: Marcellus not Mercellus

  We have corrected this spelling accordingly.

- page 7, line 20: Could you also include agricultural CH4 emissions in Figure 1?

We have added agricultural emissions to the figure.

- page 9, line 10: Mays and Cambaliza both Indianapolis.

  This is correct. One study examined CO_2 while the other focused on CH_4 (and hence we have listed the studies separately).

- page 9, paragraph beginning on line 31: A limitation is that most of these studies use data from a single campaign and provide only a snapshot of emissions. Some of the studies used tracers such as ethane to estimate contribution of landfills, etc. I think this is worth mentioning here.

  We have added this point to the paragraph in question.

- page 11, line 33. The verbiage "run an atmospheric transport model once per source sector" is confusing. Zhao et al. and Jeong et al. used STILT-WRF, so they generated footprints from a single WRF run. Suggest simply eliminating the phrase "once per source sector", since details of how the transport model is run may vary.

  We have eliminated this phrase accordingly in the revised manuscript.

- equation 5: x[i] not defined.

  The variable x[i] is defined in the lines of text following Eq. 3. We have added a clarifying note following Eq. 5: "All other variables are as defined earlier."

- page 12, line 28: Technicality: SCIAMACHY is not a satellite. It is the name of a sensor on the Envisat satellite.

  We have updated this line in the text accordingly.

- page 13, equation 6: A limitation of the GIM as implemented in the cited references is that the betas are spatially constant whereas in reality relationships between activity data and emissions may vary spatially or temporally.

This is true, and we have added this point to the revised manuscript. The coefficients could, in theory, be variable. For example, Fang and Michalak (2015, doi:10.1002/2014GB005034) allow the coefficients to vary from one biome to another (in the context of biospheric fluxes). Gelfand et al. (2003, doi:10.1198/016214503000170) developed a statistical model that estimates spatially variable coefficients, albeit not in the context of atmospheric inverse modeling. Either of these studies could be a starting point for implementing variable coefficients within a GIM.

- page 14, line 10: Radiocarbon measurements show that respired biogenic CO2 is significant even in winter.

  This is definitely true. However, the overall magnitude and diurnal variability of biogenic CO_2 fluxes are much lower in the winter than in summer. As a result, inverse modeling approaches stand a better chance of identifying fossil fuel flux patterns in winter than in summer (e.g., Shiga et al. 2014).

- page 15, line 15: It should be mentioned that in order for satellite measurements to be useful for understanding and tracking urban emissions, they must not only detect the presence of a large urban area but also be sufficiently sensitive to measure trends.

  This is an astute point, and we have added discussion on this point to the corresponding lines of the revised manuscript. For example, Hammerling et al. (2015) examined whether a LIDAR mission like ASCENDS would be able to detect changes in anthropogenic emissions from large regions like Europe or China.

- page 15, line 24: Limitations of the ASCENDS concept should be mentioned. For example ASCENDS will provide limited spatial coverage, infrequent revisits, and will low signal to noise for urban signatures.

  Good suggestion. We agree that this information is important to mention and have added it to the corresponding lines of the revised manuscript.

- page 15, line 30: Revised launch date needed for TROPOMI.

  We have revised the launch date. The TROPOMI team currently estimates a launch date sometime in 2017 (http://www.tropomi.eu/instrument/status-0).

- page 16, line 11: More recently than what?

  We have replaced the phrase “more recently” with “Beginning in the 1940s, ...”

- page 16, line 22: The description of current radiocarbon sampling could be improved. More specificity is needed, especially regarding the temporal density of samples in the current network compared to what is recommended by Basu et al.

  We have added more specific information on the available radiocarbon observations at tall tower and aircraft sites in the US.

- page 17, line 2: Impact of disequilibrium fluxes on estimated emissions can be mitigated if major urban areas have both upwind and downwind sampling.

  Good point. We have added this information into the corresponding lines of the revised manuscript.

- page 17, line 17: Typo “now markets and”.

  We have fixed this typo in the revised manuscript.

- page 17, line 18: Detlev Helmig’s lab at INSTAAR has been measuring ethane in whole air samples from the NOAA global network for many years (http://www.nature.com/ngeo/journal/v9/n7/abs/ngeo2721.html). There is a also a new instrument that is now being used to measure ethane from whole air samples North American tall towers and aircraft.

  We have updated these lines of the revised manuscript to reflect this information.

- page 18, lines 15-20: Repeated use of “far more”. Not quantitative.

  We have replaced the words “far more” in the corresponding paragraph.

- page 18, line 25: Perhaps should point out that intensive measurement campaigns provide only a snapshot and, unless repeated, provide no information about how emissions may vary over time.

  This is a great point, and we have added it to the corresponding lines of the revised manuscript.

- page 19, line 13: I don't think it is helpful or fair to single out Environment Canada for criticism (especially since focus of this review is US emissions), though your general point about data not being readily accessible is valid. CO2 data from Environment Canada through 2015 is available from the GLOBALVIEWplus_v2.1 ObsPack available here (http://www.esrl.noaa.gov/gmd/ccgg/obspack/data.php). Hopefully a similar product will be available soon for CH4.

  We have removed this reference from the revised manuscript.

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Thank your for the suggested references; these are very helpful. We have added them in to the revised version of the manuscript.

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Discussion paper

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

Scot M. Miller[1] and Anna M. Michalak[1]

[1]Department of Global Ecology, Carnegie Institution for Science, Stanford, CA, USA

Correspondence to: Scot M. Miller (scot.m.miller@gmail.com)

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 the 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~~ mixing ratios to infer emissions at the surface; thus far, these efforts have had ~~little success identifying~~ some success identifying urban CO_2 emissions ~~from anthropogenic sources but~~ and 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.

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 pledged to decrease its GHG emissions by 26–28% relative to 2005 levels by 2025 as part of the Paris Agreement negotiated at COP21 (UNFCC) . In parallel with this agreement, the US Environmental Protection Agency (EPA) ~~recently announced~~ has finalized CO_2 and CH_4 emissions regulations for numerous source sectors ~~as part of~~ under 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. 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. 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~~ Many 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. The new presidential administration that assumed office in January 2017 has announced its intention to discard the plan.) (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~~ May of 2016, EPA issued a rule that ~~would~~ will decrease CH_4 emissions from oil and gas operations by 40–45% relative to 2012 levels ~~(US EPA, 2016a) .~~ by 2025 (US EPA, 2016c) . In August of 2014, the US EPA, US Department of Agriculture (USDA), and US Department of Energy (DOE) released the *Biogas opportunities roadmap* targeting voluntary reduction strategies for agriculture (USDA et al., 2014) . Last but not least, ~~the EPA announced proposed~~ EPA announced 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~~ July 2016 (EPA, 2016b) . It is important to note that a number of these national policies are implemented at the state level. ~~US federal policies like the Clean Power Plan are implemented through plans devised by each state ; each state~~ For example, each state has a different

emissions reduction target under the Clean Power Plan, 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~~ mixing ratios 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 emissions – frameworks that can synergistically leverage the information content of bottom-up datasets ~~, atmospheric GHG data,~~ and top-down strategies using atmospheric GHG data. This review paper focuses on these opportunities.

~~Future efforts to synthesize these strategies~~ These frameworks 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 trend to trends in specific source sectors. 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~~ became widely used 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~~ Biospheric and fossil fuel sources 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~~ These sources are often co-located and trends in one could be mistaken for ~~a trend in~~ trends in the other. In addition, future changes in biospheric CO_2 and CH_4 sources may be natural or human-caused ~~GHG emissions (or vice versa).~~ (e.g., land use change, emissions induced by climate change, biological and/or geological carbon sequestration). Disentangling these natural and human causes will be challenging. Note that GHG fluxes from the biosphere and biological/geological carbon sequestration are beyond the scope of this review.

In this article, we explore ~~these challenges~~ the challenge of estimating sector-specific emissions 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; 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)~~ (e.g., P and global scales ~~(e.g., Rayner et al., 2010; Andres et al., 2011; Oda and Maksyutov, 2011; Olivier et al., 2014)~~ (e.g., Rayner et al., 2010; . 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, 2016b): 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. According to EPA, "Because this energy of the synthetic natural gas is already accounted for as natural gas combustion, this amount of energy is deducted from the industrial coal consumption statistics to avoid double counting" (US EPA, 2016c, p. A-31) . 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, 2016b).

IPCC guidelines also require a reference approach: an additional verification or consistency check against fuel production, imports, and exports (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~~ similar to 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~~ lights at night) 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~~ Emissions in the latter study are within 8.5% of Federal Highway Administration fuel consumption statistics but ~~that~~ differ from the commonly-used, global-scale EDGAR inventory (Olivier et al., 2014; European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Age 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~~ McDonald et al. 's 2014 emissions estimates differ from EDGAR by 20-80% at the municipal level, though the two inventories produce nearly identical national totals.

At the 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 ~~.~~ for the year 2002. 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 (available for 1970–2010) has moved from a $1^\circ \times 1^\circ$ lat/lon resolution to $0.1^\circ \times 0.1^\circ$ ~~(Olivier et al., 2014)~~ (Olivier et al., 2014; European Commission, Joint Research Centre (JRC)/Netherlands Environmen In a separate effort, Andres et al. (2011) estimated CO_2 emissions for 80 countries for the years 1950–2006 with a particular focus on estimating the seasonal cycle of CO_2 emissions.

A number of studies have also ~~leveraged more rigorous~~ incorporated more detailed activity data and EFs to estimate anthropogenic CH_4 emissions ~~.~~ at both regional and national scales. At the regional scale, Jeong et al. (2014) and Lyon et al. (2015) ~~estimate~~ estimated oil and gas CH_4 emissions ~~for California~~ from California for 2010 and the Barnett Shale region for 2013, respectively. Both studies 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~~cattle, 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~~ larger emissions more in line with existing top-down estimates. At the national scale, Maasakkers et al. (2016) created a gridded version of EPA's CH_4 inventory (0.1 × 0.1 lat-lon, monthly resolution for 2012). Maasakkers et al. (2016) point out that the spatial distribution of their estimate is different from EDGAR, particularly for the oil and gas industries. Oil and gas emissions in EDGAR correlate with population density while emissions in Maasakkers et al. (2016) are concentrated in drilling basins.

A number of additional studies also ~~leverage~~ employ 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~~ satellite images of lights at night. 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~~ used datasets like population density~~and economic activity as inputs into their model, constrain or fit their emissions model using nightlight data, and~~ , carbon intensity of energy, and satellite images of lights at night, and they reported national emissions totals. Davis and Caldeira (2010) used a very different approach from any of the above studies. The authors ~~build~~ built a CO_2 inventory based upon economic imports and exports and ~~explore~~ explored 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~~ incorporate 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~~ 25,000 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~~ 3,439 homes or 5,263 cars (EPA, 2015). The agricultural sector is excluded from this threshold and is not required to report its emissions. Despite ~~this omission~~these omissions, 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 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~~ mixing ratios 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~~ of CH_4 emissions from manure.

On-road measurements provide a picture of emissions that is one spatial scale larger than direct facility observations. This strategy usually entails measuring trace gas mixing ratios from a ground-based vehicle either on public roads (e.g., Maness et al., 2015) or private roads in partnership with the facility owner (e.g., Roscioli et al., 2015) . 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~~ facility-level, continuous emissions monitoring; it is typically only used for large point sources ~~, and are generally not used in existing inventory estimates~~ like power plants (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~~ and 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, 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~~ leakage rates – from 0.3% in Pennsylvania's ~~Mercellus~~ Marcellus 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 (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 the 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~~ 1.2 to 1.9 ~~(Jeong et al., 2013; Wecht et al., 2014b)~~ (Jeong et al., 2013, 2016; 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 ∼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~~uncertainty bounds. 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~~ uncertainties of ±65% (Beusse et al., 2014). Beyond the oil and gas industry, Owen and Silver (2015) also argue that many EFs for agriculture ~~are insufficient~~too low. These estimates are based upon a small number of pilot or lab experiments that were not explicitly designed for GHG inventory development.

3 Top-down, inverse modeling strategies

In this section, we discuss inverse modeling strategies – strategies that leverage observations of atmospheric GHG ~~concentrations~~ mixing ratios 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~~levels. These efforts use observation networks that are sensitive to emissions across broad geographic regions~~, but these~~. 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~~ mixing ratios using an emissions inventory and an atmospheric transport model. Subsequently, ~~one can~~ these studies scale the inventory ~~such that modeled concentrations reproduce measured atmospheric concentrations~~using a single scaling factor (β) to better match modeled mixing ratios against measured mixing ratios:

$$\mathbf{y}y_k = \underline{H}\sum_{j=1}^{m_s \cdot m_t} h_{j,k}(\mathbf{x}^a x_j^a) + \underline{\epsilon}\epsilon_k \tag{1}$$

$$\mathbf{x}^a x_j^a = \beta \mathbf{x}^b x_j^b \tag{2}$$

In these equations, ~~$\mathbf{y}$ is an $n \times 1$ vector of atmospheric GHG observations. The function $H()$~~ y_k is an atmospheric GHG observation at a given time and location k. It is one of n total observations ($k = 1...n$). The variable x_j denotes the emissions

from a model grid box j at a specific location and time, and the function $h_{j,k}()$ is an atmospheric transport model that relates the surface emissions ~~(**x**), ($(m_s \times m_t) \times 1$) to the observations (**y**)~~from grid box j to observation y_k. The variable m_s denotes the total 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~~ spatially and temporally (McKain et al., 2012), and in another study, the emissions varied spatially but were constant in time ($m_t = 1$) (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~~ the variable ϵ_k denotes the cumulative error in the model and measurement (e.g., ~~errors~~ error in estimated transport, in the ~~measurements~~measurement, 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 (**y**~~modeled atmospheric mixing ratios on the right hand side of Eq. 1 reproduces the n observed atmospheric mixing ratios (y_k where $k = 1...n$).

These local-scale efforts can target sources with very large emissions or very uncertain emissions. For example, ~~numerous~~ many 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 (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 instrument on the Envisat 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~~ leakage 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~~ estimated leakage 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~~ Nearly all of the oil and gas studies listed above use data from a single measurement campaign and provide a temporal snapshot of emissions. Greenhouse gas emissions reduction policies make it necessary to monitor trends, a goal that requires sustained monitoring. In addition, a locality or region must have ~~a single,~~ one 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 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 and the associated atmospheric transport errors 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~~mixing ratios. 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~~ narrowed 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 relative to box models 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. In addition, several studies have developed high resolution meteorological simulations, in part to better resolve atmospheric GHG transport in urban environments (e.g., McKain et al., 2012; Nehrkorn et al., 2013; McKain et al., 2015) .

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)~~ (Mays et al., 2009; Cambaliza et al., 2015; Lauvaux et al., 2016) . 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).

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.~~ spatially variable adjustments to existing inventories, unlike the strategies outlined in Sect. 3.1. The three strategies discussed in this section use both GHG observations and inventories to attribute sector-specific emissions. Each approach, however, ~~use~~ uses 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~~ Overall, these strategies have been relatively successful at attributing CH_4 emissions, but promising strategies for CO_2 are nascent. 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 spatiotemporal distribution of these fluxes is highly uncertain (e.g., Huntzinger et al., 2012) . These factors have limited the success of CO_2-focused efforts.

The first strategy discussed here scales the individual source sectors in a bottom-up inventory. This setup is often similar to a multiple ~~regression:~~ linear regression:

$$\mathbf{x}^a x_j^a = \sum_{i=1}^{p} \beta_i \mathbf{x}^b_i \beta_i x_{i,j}^b \quad (3)$$

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 (~~$\mathbf{y}$~~y_k where $k = 1...n$) in this approach is the same as in Eq. 1. This setup also assumes that ~~each $\mathbf{x}_i^b$ $((m_s \times m_t) \times 1$~~the initial emissions estimate ($x_{i,j}^b$ where $i = 1...p$ and $j = 1...m_s \cdot m_t$) is defined at ~~all spatial locationsand is defined for all time periods~~each of m_s spatial locations, at each of m_t time periods, and for each of p source sectors. In one study, ~~each $\mathbf{x}_i^b$~~ this initial emissions estimate was spatially but not temporally resolved (e.g., $m_t = 1$) (Zhao et al., 2009), while in another study, ~~$\mathbf{x}_i^b$~~ it was resolved in both space and time (Jeong et al., 2013). The p unknown scaling factors (~~β_i~~β_i where $i = 1...p$) 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 (~~$\mathbf{x}^a$~~x_j^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~~β_i) using Bayesian statistics; these frameworks can weigh uncertainty in the measurements

($\mathbf{y}y_k$) and in the atmospheric model ($H()h_{j,k}$) against uncertainty in the initial or prior guess for the scaling factors (This guess is typically unity.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 concentrationsmixing ratios.

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 ($\mathbf{y}y_k$ where $k = 1...n$) 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 anothereach of the p source sectors must have differing spatiotemporal patterns, and each column sector must explain substantial variability in ythe observations (y_k). If the former condition does not hold, then the individual source sectors $\mathbf{x}_i^b$ are some of the p source sectors will be collinear; colinearity can lead to unphysical scaling factors ($\beta_i\beta_i$ where $i = 1...p$) 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 scales an emissions inventory at the model grid level to better reproduce the atmospheric observations ($\mathbf{y}y_k$ where $k = 1...n$). All of the strategies discussed previously scale the spatial patterns in an existing inventory. By contrast, this strategy estimates an emissions level for scales the emissions level at each location in the model domain, and the resulting estimate (x_j^a where $j = 1...m_s \cdot m_t$) 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. (x_j^b). These estimates have the following general form:

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

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, Note that x_j^b and $\otimes$ is a Kronecker product that repeats the vector of x_j^a are the total emissions from model grid box j, not the emissions by sector. Hence, the 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. β_j where $j = 1...m_s \cdot m_t$) adjust total emissions, and all of the $m_s \cdot m_t$ factors are typically estimated simultaneously. Several studies estimate scaling

factors that vary spatially but are the same at each time step (e.g., Wecht et al., 2014a, b; Turner et al., 2015) . One study allows the scaling factors to vary in both space and time (Jeong et al., 2016) . 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~~the scaling factors, particularly when ~~the scaling factors (β)~~ these factors are under-constrained by the available observations (~~$\mathbf{y}$~~y_k where $k = 1...n$) (e.g., Rayner et al., 2016).

This ~~approach~~ strategy 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,~~ , but several studies have adapted this ~~strategy to support sector-specific~~ approach for source 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~~ emissions in each model grid cell using the attribution in a bottom-up ~~inventory:~~ inventory. For example, let's say that an inventory estimates that 60% of the emissions in a given grid cell are from oil and gas and 40% are from cattle and manure. The inverse modeling estimate will attribute emissions in that grid box in the same proportion:

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

All variables in this equation are as defined earlier. As a result of this setup, the total emissions in any one model grid box may differ from the inventory. However, 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) and Jeong et al. (2016) leveraged this strategy to estimate CH_4 emissions for California using ~~measurements from the CALNEX aircraft campaign~~aircraft and tower-based observations, respectively. 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~~Envisat/SCIAMACHY and the GOSAT satellite, 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. 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)~~ (Miller et al., 2013, 2016; Shiga et al., 2
This approach ~~will attribute~~ attributes 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:

$$\mathbf{x}^a x_j^a = \sum_{i=1}^{p} \beta_i \mathbf{x} x_{i,j}^b + \xi_j \quad (6)$$

The ~~vectors $\mathbf{x}_i^b$~~ elements $x_{i,j}^b$ (where $i = 1...p$ and $j = 1...m_s \cdot m_t$) can be individual source sectors from a bottom-up inventory (similar to Eq. 3). The inverse model will then map the emissions ~~on to~~ onto 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 (~~$\mathbf{y}$~~ y_k where $k = 1...n$). These emissions are denoted by ~~the vector ξ ($(m_s \times m_t) \times 1$~~ ξ_j (where $j = 1...m_s \cdot m_t$), and a GIM typically labels the emissions in ~~ξ~~ ξ_j as unattributable. Furthermore, existing studies allow ~~$\mathbf{x}_i^b$ and ξ~~ $x_{i,j}^b$ and ξ_j to vary both spatially and temporally ~~(Miller et al., 2013; Shiga et al., 2014; ASCENDS Ad Hoc Science Definition Team, 2015)~~ with j, in contrast to the studies described earlier in this section. Note that existing GIM studies have fixed the coefficients (β_i) in both space and time. In reality, the relationship between $x_{i,j}^b$ and GHG emissions may vary spatially and temporally by grid box j. Two recent GIM studies have experimented with allowing the coefficients to vary by region or biome in the context of anthropogenic (Shiga et al., 2014) and biospheric (Fang and Michalak, 2015) fluxes.

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 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 $\sum_i^p \beta_i \mathbf{x}_i^b \xi_j$~~ instead of $\sum_{i=1}^{p} \beta_i x_{i,j}^b$. In the latter case, the unattributable emissions in ~~ξ~~ ξ_j indicate inaccuracies in the spatial

distribution of available inventory estimates. Existing inventories ~~do~~ did 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~~ Yadav et al. (2016) modified the existing GIM framework to better isolate anthropogenic 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~~ emissions. The authors exploited differences in the spatiotemporal properties of biospheric versus fossil fuel fluxes to do this attribution. Specifically, the authors argued that the biospheric fluxes have smooth spatiotemporal patterns, and fossil fuels emissions do not have smooth patterns. The authors then partitioned ξ_j into two components (smooth and non-smooth) and attributed these emissions to the biosphere and fossil fuels, respectively. The study examined emissions in January when biospheric fluxes are ~~small. According to Shiga et al. (2014) , the patterns in x_i^b corresponding to anthropogenic~~ smaller than in other months.

In summary, this section discuss statistical innovations that help isolate individual emissions sources. In addition to these innovations, accurate models of atmospheric transport also play a crucial rule. A number of studies indicate the deleterious influence of transport errors. For example, Shiga et al. (2014) argue that atmospheric transport errors hinder the detection of fossil fuel emissions patterns across the United States. The authors also argue that biospheric fluxes mask fossil fuel patterns to a similar degree. Numerous additional studies examine the effects of transport errors on CO_2 ~~emissions rarely explain substantial variability in atmospheric CO_2 observations.~~ modeling, though not in the context of fossil fuel emissions (e.g. Stephens et al., 2007; Liu et al., 2012; Miller et al., 2015) .

Several efforts could reduce these transport modeling errors. Like urban-scale studies (Sect. 3.1), national inverse modeling studies have also begun moving toward high resolution meteorology simulations. These studies simulate atmospheric GHG transport at high resolution over the US and Canada and utilize coarser resolutions elsewhere to save on computational costs. For example, national-scale studies using the Weather Research and Forecasting (WRF) have modeled GHG transport at resolutions up to 8–10km (Nehrkorn et al., 2010; Gourdji et al., 2012; Miller et al., 2013) , and studies using the GEOS-Chem model have simulated CH_4 transport at resolutions up to ∼50km (e.g., Wecht et al., 2014a; Turner et al., 2015) . In addition to these efforts, NASA's Atmospheric Carbon and Transport – America campaign (ACT–America, Fig. 2a) aims to diagnose and reduce atmospheric transport errors (NASA) . The campaign includes new tower sites and five years of aircraft flights across the eastern US. Many flights will travel through frontal systems and extratropical cyclones to better characterize and evaluate atmospheric transport errors.

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)~~ (e.g., Zhao et al., 2009; Jeong et al., 2012 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.,~~

~~Several satellites make total column observations of GHG concentrations: observations of CO_2~~ The current tower network is sensitive to emissions from some source sectors but not to others. Many of the NOAA tall towers and regular aircraft sites are in or near the Great Plains. As a result, the network has sensitivity to agricultural emissions and to several oil and gas basins but has little sensitivity to emissions from east coast population centers. Earth Networks, by contrast, has focused its efforts on the East Coast proximal to large population centers. The state of California has a dense network of publicly-operated towers. By contrast to these regions, the network is sparse across the western US outside of California and northern Colorado. On one hand, the population in the regions is sparse and some emissions sectors are likely to be small (e.g., ~~AIRS, SCIAMACHY, GOSAT, and OCO-2)~~vehicle emissions). On the other hand, large resource extraction regions are beyond reach of the long term monitoring network, regions like the Powder River Basin coal mining region of Wyoming or the Bakken oil and gas basin in Montana and North Dakota.

NOAA's regular aircraft monitoring network complements these tower-based sites. The flights measure GHG mixing ratios across a vertical atmospheric profile. These datasets can help evaluate vertical mixing and transport in atmospheric transport models, and observations from the middle and upper troposphere can be used to quantify background "clean air" concentrations, a necessity for the inverse modeling studies described in Sect. 3.3. A downside is that NOAA's aircraft profiles are usually limited in frequency to one or two times per month, unlike towers which often have continuous observations. Scientists at NOAA have also invented a technology known as AirCore that can observe vertical atmospheric GHG profiles from a weather balloon (Karion et al., 2010) . This technology could become a key component of the long term monitoring network in the future.

A number of intensive aircraft campaigns provide observations across entire state or multi-state regions (Fig. 2). These include the 2010 CalNex campaign (Ryerson et al., 2013) , the 2013 SEAC^{4}RS campaign (Toon et al., 2016) , and the ACT-America campaign (2015–2019) (NASA) . Few studies have used these observations to attribute state-wide emissions. For example, Wecht et al. (2014b) used CalNex data to attribute state-wide CH_4 emissions from California.

Several satellites make total column observations of CO_2 and ~~of~~ CH_4 (e.g., ~~SCIAMACHY,~~ AIRS, TES, IASI, ~~GOSAT) (Fig. 2). Streets et al. (2013) describe each~~ Envisat/SCIAMACHY, GOSAT, OCO-2, and GHGSat). Streets et al. (2013) describe a number of these satellites ~~and the respective measurement characteristics in detail~~in detail, and Jacob et al. (2016) provide a thorough overview of CH_4-observing satellites. Four of these satellites (Envisat/SCIAMACHY, GOSAT, OCO-2, and GHGSat) observe in the shortwave infrared. Relative to other satellites, these four are more sensitive to GHG mixing ratios in the lower

troposphere and, hence, to emissions at the surface (e.g., Chevallier et al., 2005; Wecht et al., 2012) . 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) .~~ (e.g., Schneising et al., 2014; Wecht et al., 2014a, b; Alexe et al., 2015; Turner 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). Wecht et al. (2014b) , however, explains that GOSAT observations are too sparse to constrain CH_4 emissions from California outside of the Los Angeles Basin.

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

This section discusses two observational strategies ~~to~~ that 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.~~ Existing satellites could hold enormous potential for estimating fossil fuel emissions. 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~~ several studies indicate that Envisat/SCIAMACHY and GOSAT should be able to constrain 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 .~~ cities or large industrial regions (e.g., Schneising et al., 2008; Kort et al., 2012; Schneising et al., 2013) . Kort et al. (2012) further argues that GOSAT could detect a trend as small as 22% from Los Angeles. OCO-2 and GHGSat should be even more capable. OCO-2 observations have a smaller footprint and precision relative to GOSAT. As a result, the satellite should be able to constrain CO_2 from large power plants (National Research Council, 2010) . The privately-funded GHGSat makes targeted observations over specific

point sources with a smaller footprint than OCO-2 and therefore should be ideal for constraining large point sources (Kramer, 2017) .

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.~~ current satellite capabilities. Keppel-Aleks et al. (2013) argue that variations in total column CO_2 due to fossil fuel emissions 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~~ retrievals of CO_2~~at a spectrometer site in Russia. Future improvements in retrieval algorithms, however, could decrease these biases~~ . Future retrieval improvements could reduce these biases (e.g., Dils et al., 2014; Buchwitz et al., 2015) . An additional challenge is that current satellites do not provide comprehensive global mapping and therefore are not well-suited for monitoring all urban areas and point sources (Fig. 2); Miller et al. (2007) point out that OCO-2 covers only 7–12% of Earth's land surface. Trend detection can also be challenging. Individual satellites have limited lifetimes, and different satellite datasets with unique error characteristics and biases can be difficult to compare.

~~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~~ Future satellites, both selected and proposed, offer a number of improvements over existing capabilities. Some, like GOSAT-2 (selected), have better precision relative to the existing generation of satellites (Matsunaga and et. al., 2016) . Other future satellites have a wide swath (CarbonSat, proposed) or are geostationary (GeoCARB and GEO-CAPE; selected and proposed, respectively). They would generate higher density observations across the US relative to OCO-2 and GOSAT (Fishman et al., 2012; Polonsk LIDAR-based missions (e.g., MERLIN and ASCENDS; selected and proposed, respectively) measure in the absence of sunlight and through thin or scattered clouds (Kiemle et al., 2011; ASCENDS Ad Hoc Science Definition Team, 2015) . As a result, these satellites would also generate dense observations relative to current satellites, particularly at high latitudes.

These future satellites should have sufficient precision and small footprints to constrain CO_2 emissions from power plants. They should also have better spatial coverage to monitor a greater number of emitters. For example, (Bovensmann et al., 2010) report that the proposed CarbonSat satellite should be able to constrain CO_2 emissions from a mid-sized power plant to within 12–36%. Other studies, by contrast, indicate that future missions like ASCENDS would have difficulty constraining regional-scale fossil fuel CO_2 emissions from the US ~~East Coast (ASCENDS Ad Hoc Science Definition Team, 2015) and could detect large~~ (ASCENDS Ad Hoc Science Definition Team, 2015) and would have limited ability to detect continental-scale 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~~ (Hammerling et al., 2015) . In addition to CO_2, future CH_4 observations 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~~ For example, the TROMPOMI sensor is schedule to launch in 2017 and should be sufficient to constrain

the largest 1% of grid cells in EPA's gridded CH_4 ~~emissions as the recent, intensive CALNEX aircraft campaign~~inventory (Maasakkers et al., 2016) , equivalent to 30% of total national emissions (Jacob et al., 2016) .

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 (^{14}C), 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~~ years before decaying back to ^{12}C (Bowman, 1990). ~~More recently~~Since the 1940s, 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~~ Radiocarbon has not been widely used, in part, because only a handful of atmospheric monitoring sites in the US report radiocarbon ~~concentrations~~measurements. An expanded observation network shows enormous potential. ~~A handful of tall tower monitoring sites~~NOAA and its partners currently measure radiocarbon in air samples from eight tall tower sites, three mountaintop sites, and four aircraft sites in the US~~report radiocarbon and only two regular US aircraft monitoring sitesdo (Basu et al., 2016) . The~~ . NOAA collects these samples up to three times per week at tall tower and mountaintop sites and collects up to two to three samples every two weeks at aircraft sites. Basu et al. (2016) explain that there were 1639 total radiocarbon measurements between July 2009 and April 2011 (21 total months). By contrast, the National Research Council (2010) recommended that the US invest $15–20 million annually to ~~build 10 radiocarbon monitoring stations across the US~~collect 5000-10000 radiocarbon observations per year, 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~~ The atmospheric abundance of ^{14}C ~~have~~ has 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 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. Atmospheric sampling upwind of anthropogenic sources could be used to characterize the biospheric ^{14}C signature and would mitigate this concern.

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~~ mixing ratios 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,~~ atmospheric observations of ethane were sparse. Research groups at UC-Irvine and NOAA have measured ethane in air samples from global background sites since 1984 and 2004, respectively (Simpson et al., 2012; Helmig et al., 2016). Each group collects samples at 40–45 sites at weekly to seasonal frequencies. Recently, NOAA has expanded its ethane measurements to its US tall tower and aircraft network. Instrumentation has also become more widely available with Aerodyne, Inc.~~now markets and~~ 's 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~~ .

The ethane content of natural gas can also 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).

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. National CH_4 ~~emissions inventories are far more uncertain than~~ inventory estimates are often uncertain by a factor of 2–3 at the sector level while CO_2 inventories ~~, and~~ typically agree to within 5% (Fig. 1). Arguably, 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). These campaigns typically provide a snapshot of current emissions and would need to be repeated in the future to estimate how emissions vary over time.

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).

Numerous future opportunities would improve scientists' ability to merge bottom-up inventories, inverse modeling, and ~~GHG concentration~~ atmospheric GHG 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~~ CH_4 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 Envisat/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~~ leakage 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 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). Recent work by Maasakkers et al. (2016) created gridded versions of EPA's activity data and represents an important step forward. 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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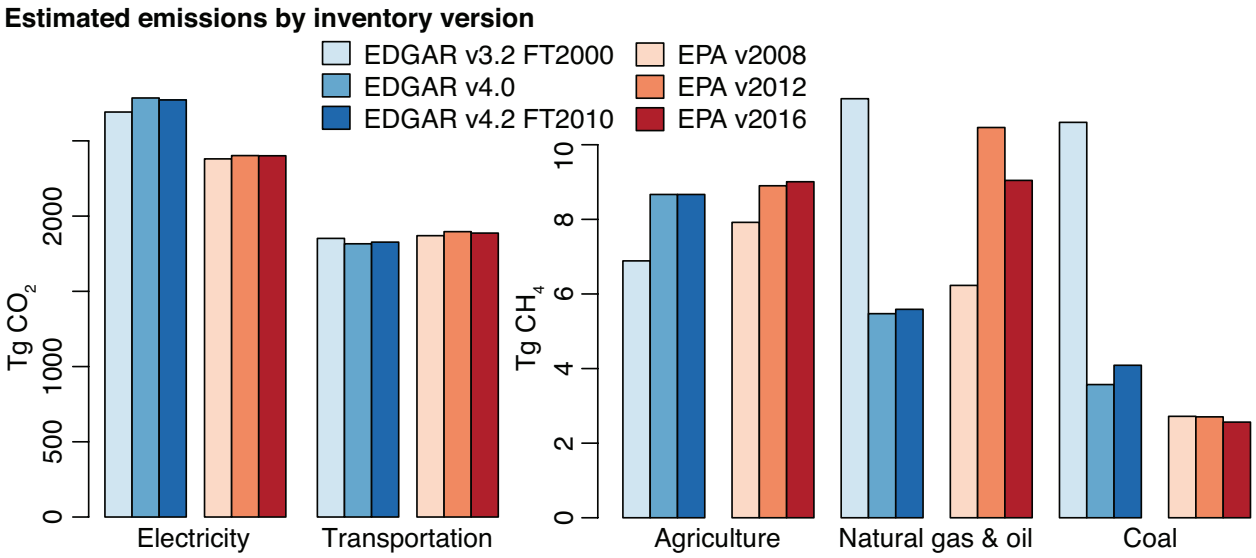

Figure 1. ~~This figure displays several~~ EDGAR and EPA inventory emissions estimates for different US fossil fuel source sectors (Olivier et al., 2014; EPA, 2016a)~~. The figure includes both the EDGAR and EPA inventories as well as~~ , including several versions of each inventory. CO_2 estimates are consistent between EPA and EDGAR and among inventory versions. CH_4 estimates, however, vary widely, an indication of uncertainty in CH_4 emissions. 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.~~

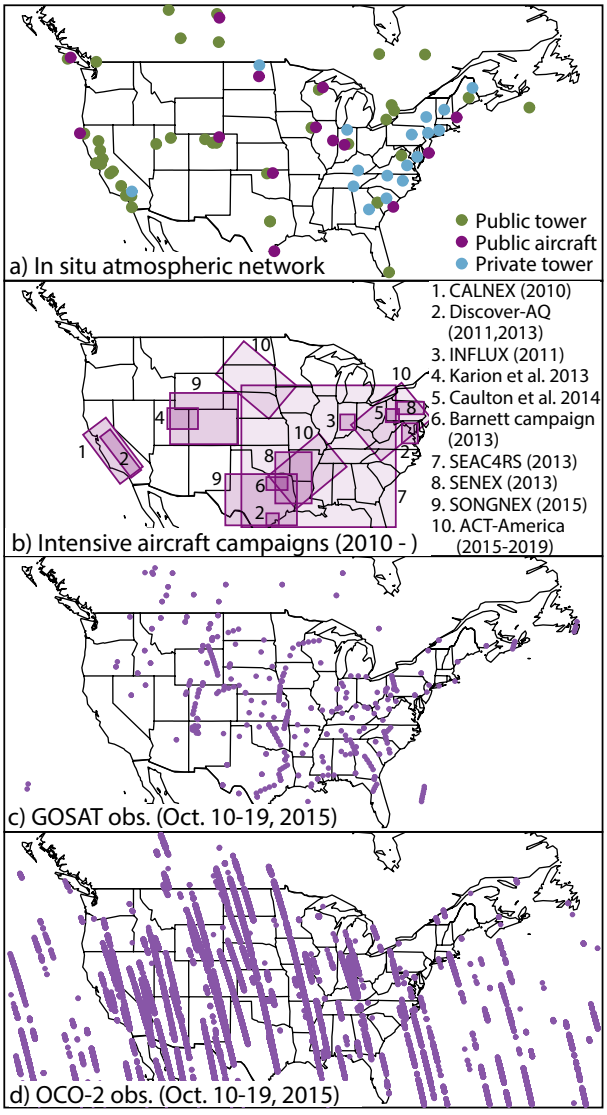

Figure 2. This figure ~~displays existing~~ highlights different CO_2 ~~observations available from~~ observation networks and how the spatial coverage of those networks differ. These networks include tower and regular aircraft sampling sites (**a**); ~~from~~ several recent, intensive aircraft campaigns (**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~~ sites in panel **a** often provides ~~far more information than an individual CO_2 total column observation from GOSAT~~ continuous or ~~OCO-2~~ (daily data while each dot in panels **c** (GOSAT) and **d** ~~, respectively~~ (OCO-2) indicates the location of a single observation. Public towers and public aircraft sites are operated by NOAA, DOE, Environment Canada, and partners~~.~~, and the sites shown are current through 2016. Private towers are operated by Earth Networks~~.~~, and the locations here are current through 2012. Most tower and aircraft sites also include CH_4 observations.

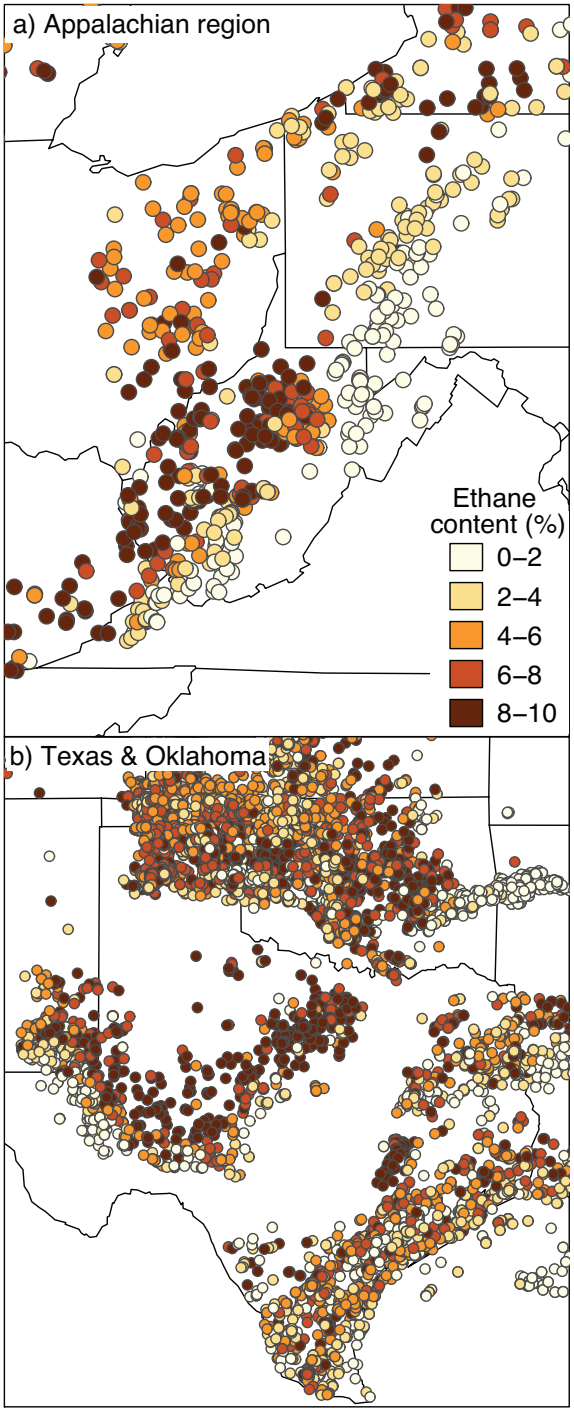

Figure 3. ~~Ethane~~ This figure shows the variability in ethane content of natural gas ~~samples from~~ for two major drilling regions of the ~~USGS Geochemistry Laboratory Database (USGS Energy Resources Program, 2015)~~ United States. Ethane content is a key parameter when estimating oil and gas CH_4 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). All data in this figure are from the USGS Geochemistry Laboratory Database (USGS Energy Resources Program, 2015) .