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Comparisons of Airborne Measurements and Inventory Estimates of Methane Emissions in the Alberta Upstream Oil and Gas Sector

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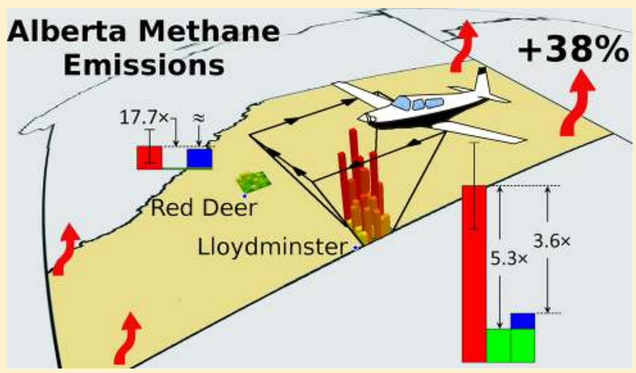
Supporting Information

ABSTRACT: Airborne measurements of methane emissions from oil and gas infrastructure were completed over two regions of Alberta, Canada. These top-down measurements were directly compared with region-specific bottom-up inventories that utilized current industry-reported flaring and venting volumes (reported data) and quantitative estimates of unreported venting and fugitive sources. For the 50 × 50 km measurement region near Red Deer, characterized by natural gas and light oil production, measured methane fluxes were more than 17 times greater than that derived from directly reported data but consistent with our region-specific bottom-up inventory-based estimate. For the 60 × 60 km measurement region near Lloydminster, characterized by significant cold heavy oil production with sand (CHOPS), airborne measured methane fluxes were five times greater than directly reported emissions from venting and flaring and four times greater than our region-specific bottom up inventory-based estimate. Extended across Alberta, our results suggest that reported venting emissions in Alberta should be 2.5 ± 0.5 times higher, and total methane emissions from the upstream oil and gas sector (excluding mined oil sands) are likely at least 25−50% greater than current government estimates. Successful mitigation efforts in the Red Deer region will need to focus on the >90% of methane emissions currently unmeasured or unreported.



INTRODUCTION

The Government of Canada has proposed new regulations intended to deliver on its "commitment to reduce emissions of methane from the oil and gas sector by 40−45% below 2012 levels by 2025".[1] The Province of Alberta is Canada's largest producer of fossil fuel resources, in 2015 accounting for 68% of Canadian natural gas production,[2] 47% of light crude oil production, and 80% of all crude oil and equivalent production (i.e., crude oil, synthetic crude oil, crude bitumen, condensate, and pentanes plus).[3] Alberta has separately announced plans to develop regulations to reduce methane emissions in the oil and gas sector through a combination of new design standards, improved measurement and reporting, and regulated standards.[4] As of this writing, draft federal regulations are under review, while Alberta's proposed regulations are under active development. Especially important questions for the success of these regulations include the accuracy of the assumed baseline methane emission estimates, the accuracy and completeness of current reporting, and the nature and distribution of sources.

Uncertainty in true methane emission magnitudes, especially from unreported and fugitive sources, complicates the identification and implementation of the most effective methane mitigation options. Several recent studies have highlighted this challenge, where impacts of vented and leaked methane can significantly increase the effective carbon intensity of one fuel source relative to another.[5−9] While much of the recent focus in the literature has been on determining methane emissions associated with hydraulically fractured natural gas production,[10−13] methane emissions are an important concern across the entire upstream oil and gas sector.[14−16]

To date there have been few measurement studies of methane emissions from oil and gas developments in Canada. A notable exception is a very recent mobile survey study of

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natural gas developments in northeastern British Columbia, Canada, which found that ~47% of active wells emitted detectable methane.[17] Another recent study[18] focused on collecting detailed activity data (i.e., counts of pneumatic equipment and numbers of leaks) across different regions in Alberta. Moreover, some resources and operating practices in the Canadian energy sector are unique in North America. Apart from oil sands development, this includes production of heavy oil resources in the Lloydminster region on the border between Alberta and Saskatchewan as well as the Peace River area of Alberta. Based on our analysis of production data as further discussed below, we estimate that in 2016, heavy oil accounted for 33% of conventional oil production in Alberta (i.e., excluding oil obtained through mined oil sands production).

The objectives of this study were to (i) generate an up to date, spatially resolved, bottom-up inventory-based estimate of methane emissions from the Alberta upstream oil and gas sector following the approaches used in the Canadian national inventory while incorporating current well- and facility-level volumetric and activity data for 2016 as reported by industry to the Alberta Energy Regulator (AER), (ii) quantify regional methane emissions using airborne techniques in two distinct oil and gas producing regions of Alberta, and (iii) directly compare these top-down and bottom-up methane emissions estimates. The methodological comparison has implications both for understanding the accuracy and completeness of current emission reporting, and for evaluating the effectiveness of current federal and provincial regulatory efforts aimed at reducing methane emissions in this sector.

CURRENT EMISSION INVENTORIES AND REPORTING

Official inventory estimates of greenhouse gas (GHG) emissions in Canada are provided in Environment and Climate Change Canada's (ECCC) National Inventory Report (NIR).[19] Based on the current ECCC inventory for 2014 (most recent data available), just over one-quarter (26%) of Canada's total GHG emissions and 44% of Canada's total methane emissions are attributed to the oil and gas sector, making it the largest source of both total GHG and methane emissions in Canada. In Alberta, approximately half of the province's total GHG emissions, and 70% of its total methane emissions, are from the oil and gas sector. Further analysis of underlying ECCC methane emissions inventory data are shown in Figure 1, estimated by production type and general source category. These are the specific emissions to be addressed by proposed federal and provincial regulations.

As shown in the left panel of Figure 1, the majority (88%) of the ECCC estimated 1.26 Mt of methane emissions in the Alberta oil and gas sector is from upstream natural gas and "conventional" oil production, with the remainder coming from oil sands mining and upgrading (11%) and downstream refining and distribution (0.6%). The latter contributions from mined oil sands operations are generally diffuse and difficult to quantify, and include fugitive methane released during large-scale surface mining[21] as well as biogenically generated methane emitted from tailings ponds.[22−24] Because practical options for mitigating these diffuse sources are understood to be limited, proposed Alberta regulations for oil sands have focused on setting a future cap on total greenhouse gas emissions of 100 $MtCO_2e$, leaving approximately 30 $MtCO_2e$ of "room" for continued growth beyond current emissions levels.[25] Methane emissions from oil sands mining

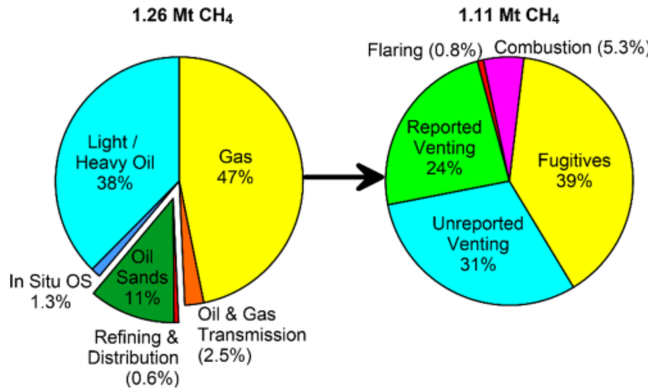

Figure 1. 2014 Methane emissions from the oil and gas sector in Alberta as derived from ECCC's National Inventory Report.[20] The left panel highlights contributions from different production types; the right panel distinguishes the emission types from natural gas and conventional oil production (i.e., excluding mined oil sands). Mt = million metric tonnes, equivalent to one teragram (Tg),

operations are also notably excluded from the proposed Federal regulations. Thus, successful cuts in overall methane emissions will primarily need to be achieved through reductions from sources outside the mined oil sands.

The right panel of Figure 1 shows the source breakdown of the 1.11 Mt of methane emissions from upstream oil and gas production (excluding mined oil sands), as derived from the current ECCC inventory. A critical observation is that only one-quarter (24.8%) of the Alberta methane emissions in the official inventory is from directly reported data. This fraction is ultimately derived from whole-gas venting and flaring volumes from active oil and gas facilities reported by industry to the Alberta Energy Regulator (AER) in accordance with AER Directive 60.[26] It is important to note that the greenhouse gas emission factors for crude oil and crude bitumen presented by AER in ST60B[36] are based only on these reported data, and do not include the other ~75% of unreported source emissions as estimated in the ECCC inventory.

Furthermore, although oil and gas operators ("industry") in Alberta are required to submit reportable flared and vented whole gas volumes to AER on a monthly basis, source data used in the federal ECCC inventory are only fully updated approximately every five years, most recently in 2014 using baseline data for 2011.[27] In interim years, official estimates in the NIR are generated using projections from the baseline year, based on activity data available to ECCC (which are generally less detailed than those maintained by AER in their general well file and related production accounting data).[28] Thus, the methane attributed to reported venting and flaring in the ECCC 2014 inventory data is actually scaled from 2011 reported data, where the methane component of these volumes is calculated using assumed average gas compositions for different types of wells and fixed flare efficiencies of 98%.[29]

Estimates for the remaining three-quarters of methane emissions in Figure 1 that are not traced back to reported volumes—broadly categorized as fugitive emissions, unreported venting, and methane emissions from combustion sources—are derived from a combination of emission factors and reported or estimated activity data (e.g., numbers of drilled oil or gas wells; assumed typical numbers of pumps and vessels per site based on field survey data; etc.).[29] These unreported venting sources "may include instrument vent gas, compressor start gas, purge

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gas and blanket gas that is discharged directly to the atmosphere, dehydrator still column off-gas" etc. that is "not normally included in reported vented volumes".[29] Fugitive sources in the inventory include leaking equipment such as valves or compressor seals, tank/truck loading and unloading operations, storage losses, and accidental releases or spills that are similarly not included in currently reported data.

■ MATERIALS AND METHODS

Measurement Regions. Figure 2 shows the geographic distribution of industry reported vented whole gas volumes in the province of Alberta in 2016. As further detailed below, Figure 2 is based on raw monthly production accounting data submitted by industry to AER, and obtained directly from AER. All upstream oil and gas facilities (i.e., oil and gas batteries, gas plants, gas gathering systems, etc., but excluding mined oil sands operations) were included in the data set. The actual reported volumes shown in Figure 2 should closely correspond to the green 24% "reported venting" slice of Figure 1 as estimated by ECCC. Reported venting is heavily concentrated in the Lloydminster region of the province, and is associated with heavy oil production in that area. Except for some elevated venting east of Peace River that is also associated with heavy oil production and isolated activity near Grande Prairie, the remainder of the province shows generally uniform, and about an order of magnitude lower venting levels.

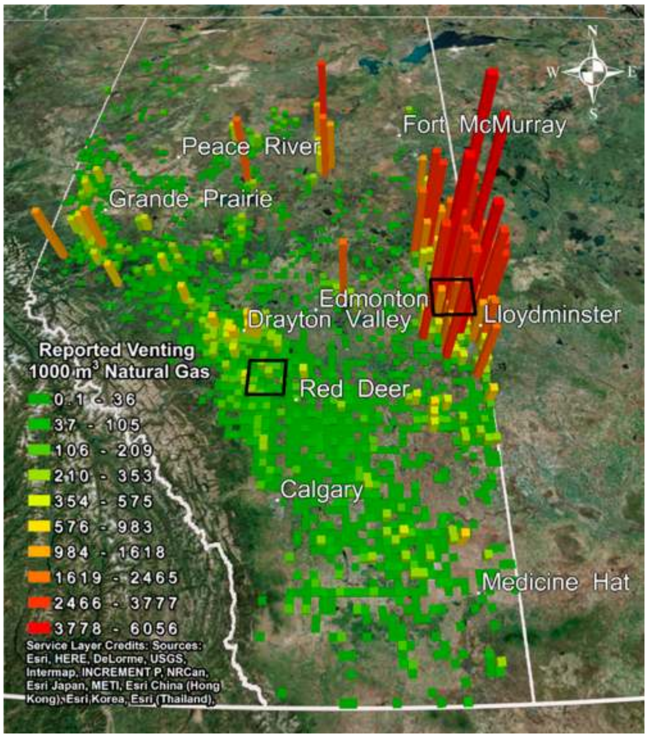

Figure 2. Geographic distribution of industry reported venting volumes in Alberta in 2016. Selected measurement regions are indicated with black squares.

Two contrasting measurement regions were defined—one near Lloydminster and one near Red Deer—as indicated by the black squares on Figure 2. The Lloydminster region was dominated by heavy oil production, whereas the Red Deer region was characterized as a mix of older natural gas and light oil production. As illustrated in Figure 3, these regions were selected considering several parameters, including magnitudes of reported venting, types of oil and gas facilities within the regions, local density of oil and gas wells, and presence/absence of other industrial facilities identified using the National Pollutant Release Inventory (NPRI) database.[30] Although methane emissions are not included in the NPRI, the database is still useful for identifying types and locations of industrial facilities not associated with oil and gas that emit any one of more than 300 species of interest (e.g., NO_x, PM, CO, VOCs, 212 listed substances of interest, 30 individual PAH species, and 7 dioxin and 10 furan/hexachlorobenzene species) above required ECCC reporting thresholds. In practice, this includes a wide range of operations including waste treatment facilities and mine sites.

The Lloydminster measurement region (Figure 3a,b), was selected to capture the region of highest reported venting in the province. The larger area (60 × 60 km), as compared to the Red Deer region (50 × 50 km), was chosen to ensure well-defined boundaries based on the distribution of active wells. Overall there were 2291 heavy oil wells (identified as producing from a deposit rather than a pool), 214 gas wells, 0 gas plants, and one in situ oil sands battery/injection facility within the measurement region. Most, if not all, of these heavy oil facilities would be expected to be characterized as CHOPS (cold heavy oil production with sand)[31] facilities. This type of production is noteworthy in that it frequently involves venting of methane from the production casing directly to atmosphere.[32] Among the thousands of oil and gas facilities within the final defined region, there was one NPRI reporting facility not associated with oil and gas (a salt production facility), which is not expected to be a source of methane.

The 50 × 50 km Red Deer measurement region (Figure 3c,d), reported venting levels typical of much of the province. The density of oil and gas sites was extremely high, and all NPRI reporting facilities contained within the selected region were associated with oil and gas production. Overall the Red Deer region included 2053 gas wells, 613 oil wells, and 11 gas plants.

Regional Bottom-Up Inventory Calculations. Bottom-up inventory estimates were generated starting from raw monthly production accounting data submitted by industry to Petrinex, a production accounting system used for regulatory reporting and royalty calculations. Petrinex is jointly governed by the provinces of Alberta and Saskatchewan, and industry as represented by the Canadian Association of Petroleum Producers (CAPP) and the Explorers and Producers Association of Canada (EPAC). These data, obtained in collaboration with AER, parallel publicly available facility production information sold in the AER Products and Services Catalogue. However, the form of the data obtained allowed volumes reported at individual batteries (i.e., upstream facilities where raw effluent—a combination of gas, water, and/or oil—from one or more wells is initially collected, separated for measurement and sometimes pretreated) and other facilities to be linked back to individual producing wells, which was critical for many aspects of the spatially resolved inventory development. These facility-level volumetric data were linked with detailed well activity data available in AER's general well data file, which allowed identification of types of wells feeding into batteries. In particular, this allowed venting volumes reported in aggregate at "paper batteries" (i.e., groups of disperse, physically disconnected wells reporting aggregated volume data as if they were connected at a single battery, as mostly occurs within the

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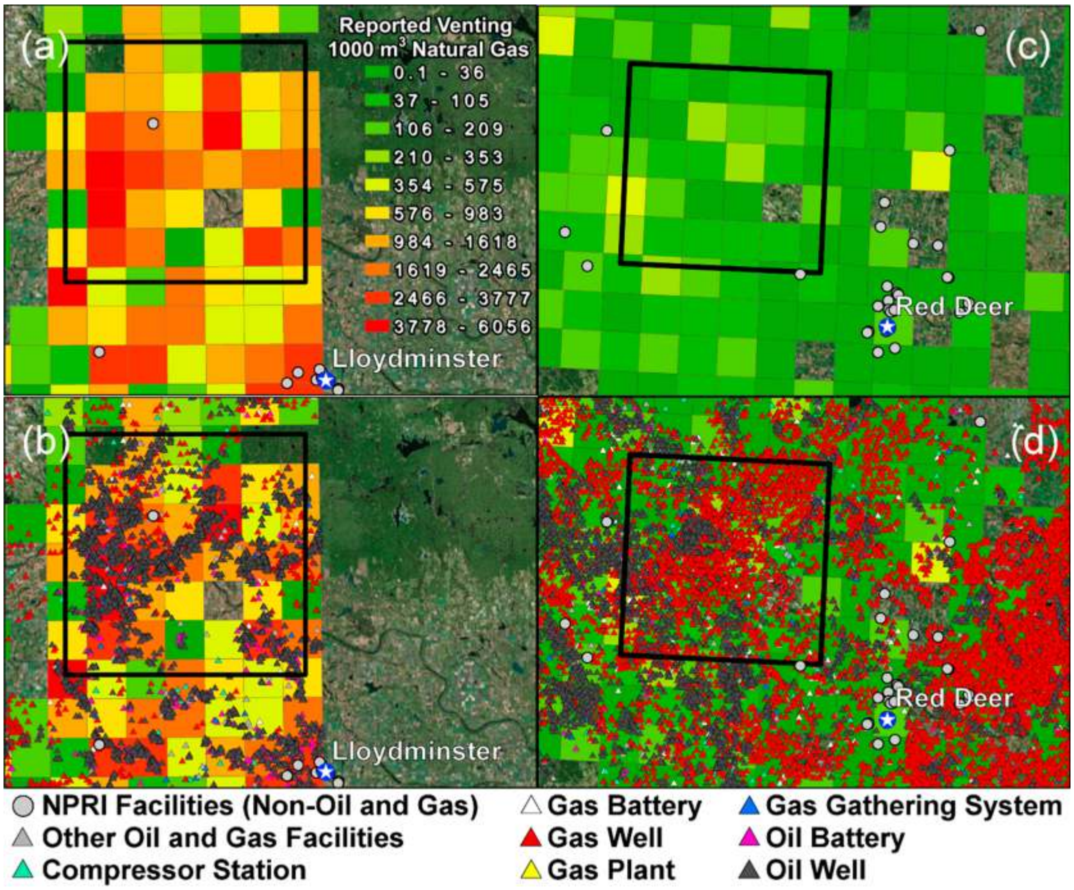

Figure 3. Measurement regions of interest near the cities of Lloydminster and Red Deer in Alberta. The background contour grid shows the local magnitudes of reported venting using the same color scale as Figure 2. Gray dots added in (a) and (c) show nearby nonoil and gas industry facilities appearing in the National Pollutant Release Inventory (NPRI). Colored triangles appearing in (b) and (d) indicate oil and gas wells, oil and gas batteries, gas plants, compressor stations, gas gathering systems and other associated upstream oil and gas facilities. Background satellite imagery source layer credits: Esri, DigitalGlobe, GeoEye, Earthstar Graphics, CNES/Airbus DS, USDA, USGS, AEX, Getmapping Aerogrid, IGN,IGP,swisstopo, and the GIS User Community.

Lloydminster region of Alberta) to be appropriately located based on individual source wells.[33,34] Light and heavy oil batteries were distinguished based on the types of the pools or deposits from which wells feeding each battery were producing. A further distinction between heavy cold production and heavy thermal production was derived from AER-assigned battery subtypes.[35]

Under AER Directive 60, operators are required to report monthly flaring and venting (whole gas) volumes exceeding 100 m^3/month from active facilities.[26] Industry is also required to report volumes of produced gas used for onsite as "fuel"; however, since this gas may be flared (i.e., used as purge or pilot gas in a flare system), combusted (e.g., used as fuel in natural gas compressors), or vented directly to atmosphere (e.g., used to drive pneumatic equipment), these fuel gas data have limited utility in emissions estimation and are only used to estimate the small fraction of methane emitted in the exhaust of combustion systems. Monthly reported flared and vented gas volumes are submitted to AER through the Petrinex reporting system, which AER uses to produce annual summary reports of flaring and venting volumes.[36] However, industry is not currently required to report composition or methane content of vented gas. AER does not currently collect data on greenhouse gas emissions.[36]

For the present analysis, site specific gas composition data were determined starting from an AER data set of individual well gas analyses containing 312 654 useable samples associated with 117 206 well segments, each coded with a unique well identifier (UWI). Individual gas compositions at all active oil and gas batteries in Alberta were estimated using production-weighted composition data from each UWI feeding into each battery. Compositions of active UWIs without available direct gas sample data were calculated by spatial interpolation from nearby sites with data. Where possible, compositions of reported flared and vented gas at other facilities (e.g., gas plants and gas gathering systems) were similarly determined using a gas-volume-weighted average of the reported gas receipts from feeding upstream facilities, supplemented by spatial interpolation where necessary. This procedure allowed site-specific methane emissions to be calculated at individual wells, batteries, and other facilities throughout the province. Gas production-weighted methane and ethane fractions could then be accurately determined for each measurement region as further detailed in the Supporting Information (SI). Finally, the 2016 methane emissions from reported flaring and venting volumes were calculated using site-specific composition data.

As noted in the Current Inventories section, the official ECCC national greenhouse gas inventory also includes provincial estimates of emissions from unreported sources in

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Table 1. Summary of Derived Bottom-up Emissions Data within the Measurement Regions

region specific inventory data	Lloydminster	Red Deer
measurement region statistics		
GPS coordinates (centroid)	−110.517365, 53.775022	−114.433770, 52.610004
dimensions (km)	60 × 60	50 × 50
no. of active wells in 2016	2631	2672
no. of gas wells/no. oil wells/no. oil wells identified as CHOPS wells	214/2350/2291	2053/613/0
no. of new wells drilled in 2016	52	24
no. of gas/oil batteries (including single-well batteries)	42/1430	773/296
no. of gathering systems/compressor stations/gas plants/other oil and gas facilities	60/29/0/86	144/126/11/50
total volume of gas produced (10^6 m^3)	467	3511
total volume of oil and heavy oil produced (10^3 m^3)	3906	403
volume weighted mean CH_4 content of produced gas (by volume)	97.2%	82.1%
volume weighted mean C_2H_6 content of produced gas (by volume)	0.675%	7.65%
Emissions Associated with Directly Reported Sources		
industry reported venting in 2016 (1000 m^3, whole gas)	60,602	2,540
industry reported flaring in 2016 (1000 m^3, whole gas)	662	8582
CH_4 emissions from industry reported venting (tCH_4/h)	4.6	0.16
CH_4 emissions from industry reported flaring (tCH_4/h)	0.0010	0.011
combined CH_4 emissions from directly reported flaring and venting (tCH_4/h)	4.6	0.17
Estimated Additional Methane Emissions Following ECCC Inventory Methodology		
CH_4 emissions from combustion sources (tCH_4/h)	0.050	0.13
CH_4 emissions from unreported venting sources (tCH_4/h) (incl. emissions from glycol dehydrators as per Figure 4)	0.66	1.5
CH_4 emissions from unreported fugitive sources (tCH_4/h) (incl. emissions from leaks, gas migration, storage losses, etc. per Figure 4)	1.4	0.93
total estimated methane from unreported sources (tCH_4/h)	2.1	2.5
total expected bottom-up methane emissions (including reported and unreported sources) (tCH_4/h)	6.7	2.7

the oil and gas sector. Thus, to generate complete and directly comparable bottom-up inventory data for each measurement-region, estimates of unreported vented and fugitive emission sources were also calculated. This was accomplished following the same approaches used in the development of ECCC's federal greenhouse gas inventory,[28,29] while updating them to use current AER 2016 activity and production data. For each operation type (i.e., ECCC sector) and source category[28] in each region, unreported emissions were separately updated by prorating the relevant ECCC 2011 baseline-year data using currently derived, up to date and region-specific activity data. These regionalized data included numbers of existing and new wells, flared and vented gas volumes, produced gas volumes, reported fuel volumes, and produced volumes of liquids, categorized as oil, heavy oil, or crude bitumen. The end result of this analysis was a current and detailed bottom-up inventory for each measurement region that included, and separately identified, methane emissions associated with sources directly reported to AER, as well as emissions from unreported source categories listed in the ECCC national inventory.

Airborne Flux Measurements. Regional methane and ethane emission rates were calculated based on airborne measurements from a series of flights conducted during October 27 to November 5, 2016. The measurement and emission quantification methodology is detailed elsewhere[37] and is only briefly reviewed here. To estimate the magnitude of a trace gas source at the surface, a flight track consisting of a series of concentric closed paths around the source of interest is employed. These paths begin as close as possible to the ground (usually ~150 m) and climb until aircraft is well above the highest level the plume reaches before crossing the flight path, which is determined by the absence of significant upwind/downwind variability in the methane signal. The instantaneous vector flux of a target species (i.e., methane or ethane) is simply the vector wind[38] multiplied by the species density (kg m^{-3}). Density is computed using the mixing ratios reported by the flight-ready Picarro CRDS spectrometer along with the temperature from the Vaisala HMP60 probe and the pressure from the air data computer (Aspen PFD1000). At each altitude (individual closed path), the net flux into the region bounded by the path is simply the sum of the flux normal to the flight path (dot product of flux vector and flight path). The number of laps required to obtain a robust estimate varies from ~20 close to the source[37] to 1 if the aircraft is far enough downwind for the plume to mix throughout the boundary layer.[7] For the present case, the plume was well mixed, confirmed by at least one vertical profile on each lap.

The uncertainty on the measured flux for each lap considered contributions from the uncertainty in the wind measurement (~0.5 m s^{-1}), the uncertainty in the methane or ethane measurement (~1 or 5 ppb), and the uncertainty in the boundary layer height (~50 m). As further detailed in the SI, each lap was treated as an independent measurement and the variance of the fluxes among laps was used as a direct measure of the precision uncertainty. The reported 95% confidence intervals in the mean regional emissions flux considered the combined instrument and precision uncertainties, while correcting for sample size using the t-statistic.

The relative contributions of oil and gas sector and biogenic sources to these total measured methane fluxes were assessed in two ways. First, direct attribution of oil and gas sector emissions was determined using the ratio of the methane and ethane flux as measured by the aircraft. Given that biogenic sources only emit methane, the directly measured ratios, combined with knowledge of the mean methane/ethane ratio of the produced gas at local oil and gas facilities (see Table 1), allowed the mass of methane emissions attributable to oil and gas activity to be determined. For the Red Deer region, with a mean ethane

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content of 7.65%, the oil and gas attributable (i.e., fossil) component of the methane flux was estimable in this manner. However, for the Lloydminster region, where the regional ethane fraction was more than an order of magnitude lower (0.68%), the corresponding uncertainties in the measured ethane flux were more than two times the measured value, limiting the utility of this method.

In the second approach, spatially explicit (0.1° × 0.1° spatial resolution) methane emission estimates for anthropogenic sources from the Emissions Database for Global Atmospheric Research v4.3 (EDGAR)[39] were used to calculate methane fluxes from sources not associated with oil and gas (i.e., from enteric fermentation, manure, and landfills and waste) in each measurement region. Results suggest biogenic methane emissions of 0.42 tCH_4/h in the Lloydminster measurement region and 0.89 tCH_4/h in the Red Deer region. These values may be slightly conservative since the provincial total EDGAR methane emissions were noted to be 9% higher than the available total provincial methane estimates from livestock and landfills/waste in the ECCC NIR.[20] However, the EDGAR data were within the range of published CH_4 emission estimates from livestock and landfills/waste sources in U.S. oil and gas producing basins.[7,15,40−42] The potential for additional methane emissions from microbial activity in local wetlands (which are not included in the EDGAR data) was also considered using six globally gridded wetland methane emission data sets (CLM4Me,[43] DLEM,[44] LPJ-Bern,[45] LPJ-WSL,[46] ORCHID-EE,[47−50] and SDGVM[51,52]). Based on 12 consecutive years of data from 1993−2004 (latest years available), average November wetland emissions were estimated to be −0.0018 ± 0.0020 tCH_4/h in Lloydminster and 0.015 ± 0.015 tCH_4/h in Red Deer (uncertainties are standard error of the mean). The slightly negative emissions in the Lloydminster region implies that the soil sink is larger than the positive wetland emissions in November in these models. Combined with the EDGAR results, this literature analysis suggests expected total biogenic methane emissions of 0.42 tCH_4/h in the Lloydminster region and 0.91 tCH_4/h in the Red Deer region.

RESULTS AND DISCUSSION

Bottom-up Methane Emissions. Table 1 reports summary statistics and derived bottom-up inventory volumes for the two measurement regions. Although the total number of active oil and gas wells in each region was comparable, their production and methane emission characteristics were quite different. The Red Deer region contained a mix of gas (77%) and light oil wells (23%) whereas the Lloydminster region was dominated by heavy oil wells (89%) associated with CHOPS production. As shown in Table 1, average total methane emissions from reported venting and flaring volumes were 4.6 tCH_4/h in the Lloydminster measurement region and 0.17 tCH_4/h near Red Deer. Methane emissions from reported venting volumes were responsible for almost all of these totals (99.98% in Lloydminster, 93.7% in Red Deer). The significantly higher reported emissions in the Lloydminster region are indicative of the density of CHOPS production sites in the area, and their associated venting of casing gas. Using pool or deposit codes of wells feeding into batteries as criteria for identifying heavy oil production sites, CHOPS production sites accounted for 39% of all reported venting from oil production in Alberta (excluding mined oil sands).

Estimates of unreported methane sources were also generated for each region by combining ECCC baseline inventory data with up to date, region-specific volume and activity data, as summarized in the lower rows of Table 1. In combination with methane emissions derived from directly reported flaring and venting volume data as submitted to AER, this provides a more up to date and complete bottom-up inventory estimate of both reported and unreported methane sources as shown in the final row of Table 1. In the Lloydminster measurement region, methane from unreported sources added an additional 47% to that from reported venting, representing 32% of the total bottom-up methane inventory estimate in the region. In the Red Deer region, unreported sources were 15 times larger than reported sources, equating to 94% of the local bottom-up methane inventory.

This is a significant finding that affirms previous studies reporting similar challenges between inventory estimates and reporting programs in the U.S.[53] The available provincial summary data in the ECCC NIR potentially obscures the fact that in oil and gas production regions like Red Deer, methane associated with directly reported data (i.e., as currently reported to AER under Directive 60) makes up only a small fraction of total emissions. From the regional methane emission break-downs shown in Figure 4, unreported venting (e.g., pneumatic

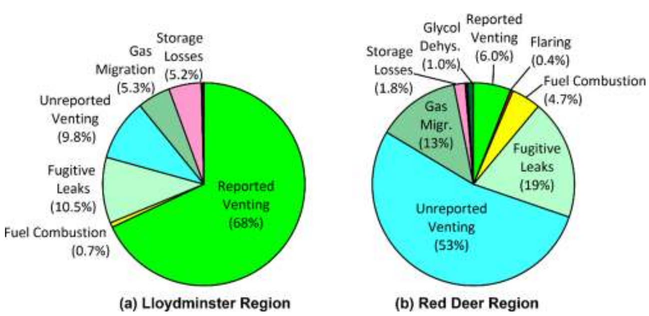

Figure 4. Relative importance of sources contributing to the bottom up methane inventory for the measurement regions near (a) Lloydminster and (b) Red Deer.

instrument vent gas, purge gas, compressor starts, tank venting, etc.) and fugitive leaks are responsible for nearly three-quarters (73%) of bottom-up methane emissions in the Red Deer region. By contrast, the bottom-up inventory suggests that the Lloydminster region is dominated by reported venting (68% of methane emissions) with fugitive leaks and unreported venting as the next largest sources combining for 20%.

Regional Estimates from Airborne Measurements. Top-down measured, mean total methane emission rates were 24.5 ± 5.9 tCH_4/h in the Lloydminster region and 3.05 ± 1.1 tCH_4/h in the Red Deer region. As further detailed in the SI, the reported ranges are 95% confidence intervals about the measured mean emission rates. For the Red Deer region, the aircraft measured ethane flux of 0.53 ± 0.38 tC_2H_6/h, and the regional ethane fraction of 7.65%, implied that effectively all of the measured methane (3.07 ± 2.2 tCH_4/h) was attributable to oil and gas operations (i.e., fossil). The uncertainty range includes the estimate of 2.14 tCH_4/h that would be generated if the literature estimate of biogenic methane emissions in the region were instead assumed. However, the higher value seems more likely in the context of a recent report by GreenPath Energy Ltd.[18] inventorying pneumatic equipment and associated leaks. In a study region that almost completely overlaps the Red Deer measurement region, they estimated an average pneumatic leak rate of 6.54 tCH_4/y per well. Applying

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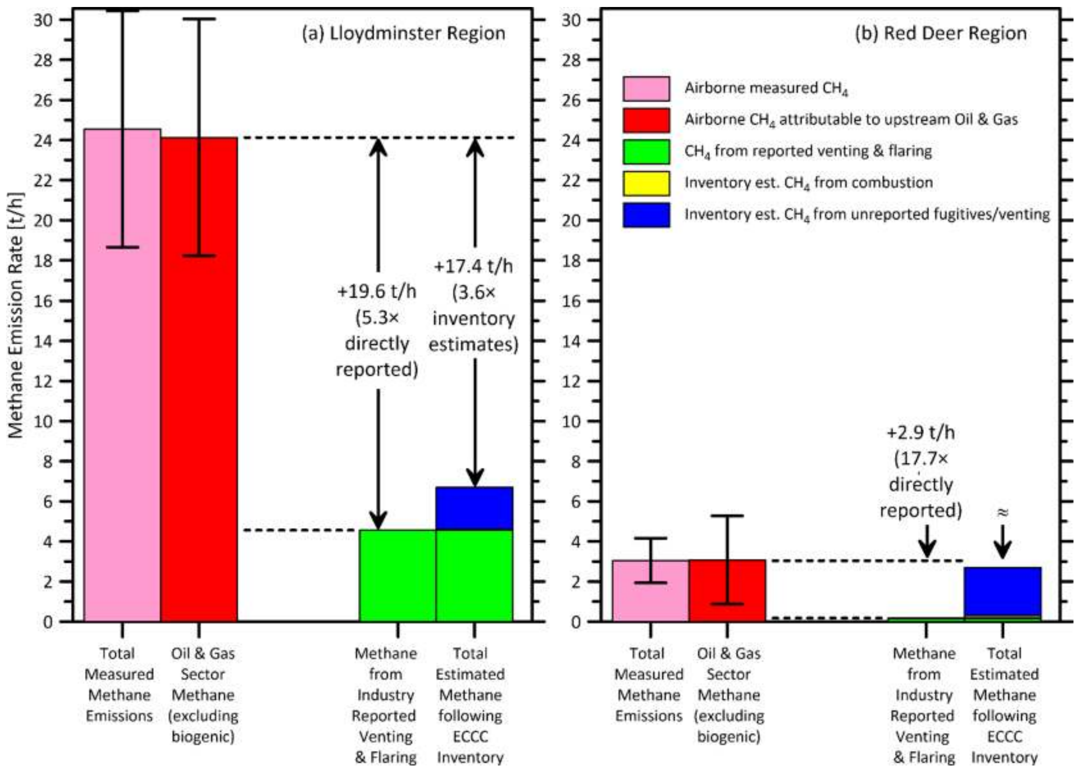

Figure 5. Top-down vs bottom-up comparison of methane emissions in the (a) Lloydminster and (b) Red Deer measurement regions.

this emission factor to the 2666 wells in Table 1 suggests a methane flux of 2.0 tCH_4/h just from pneumatic devices alone. The present aircraft based estimate suggests that other sources account for an additional ∼1 tCH_4/h.

For the Lloydminster region, oil and gas sources were similarly responsible for the vast majority of measured methane emissions, equating to 24.1 ± 5.9 tCH_4/h. Even considering the slightly larger measurement area, this is still more than five times higher than near Red Deer. Moreover, given the results for the Red Deer region, the use of the literature-based value for biogenic source estimation in Lloydminster might be considered conservative.

Top-Down vs Bottom-up Emissions Comparison. Figure 5 compares top-down measurements of methane flux in each measurement region with presently estimated bottom-up calculations as detailed in Table 1. The pink bar in each panel represents the total methane flux based on the aircraft measurements; corresponding 95% confidence intervals are indicated directly on each bar. The red bars represent the net methane flux attributable to oil and gas development activity within each measurement region, where the difference between the adjacent pink and red bars is the biogenic methane contribution. As described previously, the biogenic methane contribution for the Lloydminster region is based on EDGAR estimates, while for the Red Deer region, it is based on the directly measured ratio of ethane to methane flux.

The oil and gas sector methane emission rate in the Lloydminster region of 24.1 tCH_4/h is 3.6 times greater than the current total bottom-up inventory estimate, and 5.3 times greater than the methane emissions from directly reported venting and flaring. On an annual basis, the high rate of methane emissions alone within this small 60 × 60 km region represents GHG emissions of 5.3 $MtCO_2e$ (conservatively evaluated on a 100-year time horizon with GWP_{CH4} = 25). Using more recent IPCC GWP_{CH4} values,[54] this equates to 18/6.3 $MtCO_2e$ evaluated on a 20/100-year basis.[55] These results verify the overwhelming emission contribution of CHOPs production in this area, while further suggesting significant under-reporting or under-estimation of methane emissions to the atmosphere.

Total Red Deer methane emissions were consistent with the current regional inventory estimate once reported and unreported sources were combined, and much lower than the levels seen in the Lloydminster region. However, the aircraft measurement based methane flux was still more than 17 times greater than directly reported data would suggest, affirming the regionally derived inventory result that the vast majority of methane emissions in this area are from sources not currently monitored or reported.

Implications. In the context of proposed regulations aimed at reducing methane emissions in the Canadian oil and gas sector by 45%, large discrepancies between actual methane emissions and emissions from currently reported data present a critical challenge. With unreported emissions in regions like Red Deer accounting for 94% of the total methane emissions, the majority of reductions will need to come from sources that may not yet be identified and/or are not being measured. Specifically, assuming the source breakdown (Figure 4) in the presently estimated regional inventory for Red Deer, 70% of methane is likely to come from unreported venting and fugitive leaks. This strongly suggests a need for policies to address this reporting gap as these sources represent significant methane reductions opportunities. Research performed in U.S. fields with similar production characteristics has highlighted the presence of spatial and temporal emission patterns that require a frequent or even continuous monitoring scheme in order to control fugitive leaks.[56,57] Field measurement statistics from pneumatic equipment in particular, emphasize the importance

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of frequent inspection programs for identifying the subset of malfunctioning and high-emitting devices responsible for the large majority of emissions.[12] In addition, further empirical measurements at the site and component-level, and more comprehensive accounting of facilities, major equipment, and activity data would consistently improve bottom-up estimates.[53]

The discrepancy of a factor of 3−5 between measured methane emissions and both the reported and inventory estimates in the Lloydminster region is noteworthy, and contrasts with results for the Red Deer region, where combined reported and unreported emissions essentially matched airborne measurements. This suggests that the unexplained emissions in the Lloydminster region are attributable to unique operating practices in that area, which is characterized by a large population of CHOPS sites. The most likely source of the excess methane emissions in the Lloydminster region is underreported venting of casing gas from CHOPS sites, which is generally estimated based on the product of the measured produced oil volume and an assumed gas to oil ratio (GOR).[58] Current regulations require that the GOR be measured every six months (if the produced gas volumes are >1000 m^3/day), or as infrequently as every 3 years (if gas volumes are <1000 m^3/day).[59] One interpretation of the present results is that current measurement and reporting practices for casing gas venting via periodic GOR measurements are inadequate. This may be especially true given anecdotal data that produced gas volumes at CHOPS sites can be highly variable in time.[60] This temporal heterogeneity suggests the necessity of regular monitoring if reductions are to be achieved.

The possibility of underreporting at CHOPS sites presents important implications at the provincial level given the dominance of CHOPS region emissions in reported venting totals for Alberta (Figure 2). If the extra 17.4 tCH_4/h of methane emissions in the Lloydminster measurement region are assumed to come from CHOPS facilities, and then extended to other CHOPS production sites in Alberta while leaving current inventory estimates for all other types of facilities *unchanged*, this suggests that total reported venting in Alberta is low by a factor of ∼2.5 (range of 2.0−3.1). Relative to current inventory estimates of both reported and unreported emissions, the present results suggest that actual methane emissions from the upstream oil and gas sector (excluding mined oil sands) are likely to be at least 25−50% greater than currently estimated. Considering data gathered in other regions suggest fugitive and vented emissions are underestimated,[8,15,17,41] it seems probable that this ∼38% augmentation may be conservatively low. This also suggests further investigations would be warranted in other production regions of Alberta (e.g., Rocky Mountain House, Grande Prairie, and Peace River regions), as well as in Saskatchewan and British Columbia. Overall, the present results suggest that federal and provincial efforts to regulate methane are timely. A 45% cut in the current Alberta inventory methane emissions totals from Figure 1 implies a decrease of 500 $ktCH_4$/y. The present results suggest a reduction of 924 $ktCH_4$/y would actually be required to reach the same absolute emissions target.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b03525.

Details of the airborne measurements and regional gas compositions (PDF)

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Supporting Information

Comparisons of Airborne Measurements and Inventory Estimates of Methane Emissions in the Alberta Upstream Oil and Gas Sector

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S.1 Airborne Measurements

Airborne measurements of regional methane and ethane flux were completed during October 27 – November 5, 2016 using the quantification methodology as detailed by Conley et al.[1]. It is necessary to note the size difference in this study's measurement regions compared to that in the referenced methodology. There are important differences between measurements obtained using the Green's theorem approach for a point source and for a large field, and those differences are explored here. In general, turbulence acts to reduce concentration gradients. These gradients within a gas plume are the strongest nearest the emission source and tend to diffuse as the plume moves downwind. As a result, when measuring point sources, where an aircraft often flies within 1-2 km of the source, more measurement laps are required to reduce uncertainty as the plume has not yet mixed throughout the boundary layer. This is distinctly different when flying a large box as in the present study. In this study, the aircraft is potentially up to 50 km from the source allowing sufficient time for the plume to be well mixed when it crosses the downwind edge of the measurement path, as shown in Figure S4. Of course, the trade-off in moving further downwind is that laps take substantially longer (~1 hour for the present study), although fewer laps are required to achieve a meaningful statistical result.

Another consideration for airborne measurements of large regions is "pooling". Typically when flying a point source measurement, gas pooling from the previous night is not normally a concern. The transit time for a parcel carried by the mean wind across the flight path (~4 km) is a matter of minutes for even a modest wind. That is not necessarily the case for the size of region flown in the present study and the action of the wind before the flight must be considered. Fortunately, there is an airport at the edge of each of the boxes where winds are measured and recorded on an hourly basis. By converting the wind direction and speed to vector components and summing those hourly measurements from the night and morning before the flight, we estimated the "net" motion of an air parcel in the box. For the Red Deer field, those values were 41 km, 28 km and 62 km respectively for October 27, November 2 and 3. For Lloydminster, the values were 79 km and 116 km for November 4 and 5. These should be conservative estimates since winds generally increase with height in the surface layer and the airport only measures surface winds. Thus, pooling of emissions was not a concern in the present measurements.

S.1.1 Details of the Airborne Measurement Results and Uncertainty Calculations

Table S1 details the calculated emission rates for each lap of the measurement regions, and shows the final average emission fluxes and uncertainties, calculated as further described below.

Table S1: Summary of Top-Down emission estimates derived from the airborne measurements

Lap #	Flight date	CH_4 emission estimate [t/h]	C_2H_6 emission estimate [t/h]
Red Deer Measurement Region			
1	2016/10/27	2.91±0.52	0.26±0.27
2	2016/11/02	3.12±0.89	0.51±0.53
3	2016/11/02	2.47±0.66	0.42±0.20
4	2016/11/02	3.59±1.00	0.92±0.47
5	2016/11/03	5.32±0.70	0.77±0.78
6	2016/11/03	2.00±0.25	0.44±0.09
7	2016/11/03	1.94±0.24	0.39±0.44
	Mean Emission Rate, $\bar{x}$	**3.05**	**0.53**
	Bias Uncertainty in $\bar{x}$	**0.25**	**0.17**
	Precision Uncertainty in $\bar{x}$	**0.44**	**0.09**
	Combined Uncertainty in $\bar{x}$	**0.51**	**0.19**
	Effective Degrees of Freedom, ν_{eff}	**25**	**137**
	95% Confidence Interval in $\bar{x}$	**1.1**	**0.38**
Lloydminster Measurement Region			
1	2016/11/04	25.5±5.9	-0.047±0.05
2	2016/11/04	23.3±5.3	0.23±0.05
3	2016/11/05	25.5±5.9	0.32±0.24
4	2016/11/05	23.8±6.4	0.31±0.32
	Mean Emission Rate, $\bar{x}$	**24.5**	**0.20**
	Bias Uncertainty in $\bar{x}$	**2.95**	**0.10**
	Precision Uncertainty in $\bar{x}$	**0.58**	**0.09**
	Combined Uncertainty in $\bar{x}$	**3.0**	**0.13**
	Effective Degrees of Freedom, ν_{eff}	**2134**	**17**
	95 %Confidence Interval in $\bar{x}$	**5.9**	**0.28**

The mean emission rate for each region, $\bar{x}$, is calculated as the average of the N emission rates x_i measured for the laps of each region:

$$\bar{x} = \frac{1}{N}\sum_{i=1}^{N} x_i \tag{S.1}$$

The uncertainty in $\bar{x}$ is calculated following procedures developed by the Joint Committee for Guides in Metrology[2] and detailed by NASA[3]. The bias uncertainty in the mean emission rate is calculated from the contributing instrument uncertainties in individual lap measurements, σ_i, as follows:

$$u_{\bar{x},bias} = \left(\sum_{i=1}^{N}\left[\frac{\partial \bar{x}}{\partial x_i}\sigma_i\right]^2\right)^{1/2} = \frac{\sqrt{\sum_{i=1}^{N}\sigma_i^2}}{N} \tag{S.2}$$

The repeatability uncertainty of the measurements on separate laps is:

$$u_{\bar{x},ran} = \frac{s_x}{\sqrt{N}} \quad \text{(S.3)}$$

where s_x is sample standard deviation of the measured emission rates in the different laps.

The combined bias and repeatability uncertainty is then:

$$u_{\bar{x}} = \sqrt{u_{\bar{x},bias}^2 + u_{\bar{x},ran}^2} \quad \text{(S.4)}$$

The effective degrees of freedom, ν_{eff}, for the combined uncertainty comes from the Welch-Satterthwaite formula [2,3]:

$$\nu_{eff} = \frac{u_{\bar{x}}^4}{\frac{u_{\bar{x},bias}^4}{\nu_{bias}} + \frac{u_{\bar{x},ran}^4}{\nu_{ran}}} = \frac{\nu_{ran} u_{\bar{x}}^4}{u_{\bar{x},ran}^4} \quad \text{(S.5)}$$

The degrees of freedom for the repeatability uncertainty, ν_{ran}, is the number of laps contributing to the final average measurement minus 1. The degrees of freedom in the bias uncertainty, ν_{bias}, may be considered infinite based on the number of integrated measurement during a lap. Finally, the confidence interval about the mean emission rate may be calculated as:

$$\bar{x} \pm t_{\alpha/2,\nu_{eff}} u_{\bar{x}} \quad \text{(S.6)}$$

where $t_{\alpha/2,\nu_{eff}}$ is the t-statistic based on the effective degrees of freedom, calculated here at 95% confidence with $\alpha/2 = 0.025$.

Figure S1 – Figure S4 show a series of maps and plots that illustrate the data gathered during the flights, with one flight in each region serving as an example. Vertical profiles were conducted throughout each day to ensure the boundary layer was well mixed, and to identify the height of the boundary layer. Given the time of year (November) and the high latitude (~54°), the boundary layer top was often below the minimum flight altitude (150 m AGL). Consequently, flight legs were scrutinized for periods when the airplane ventured out of the boundary layer, and those legs were excluded from calculations as further detailed in Section S.1.2 below. For the first Red Deer flight, 4 total laps were flown, three of which were excluded because of excursions above the ABL leaving one lap to be used in the final analysis. At attempt was made on 10/30/2016 but fog prevented the aircraft from flying within the ABL on any leg. The Red Deer flight on 11/2/2016 included 4 total laps with one excluded, leaving 3 in the final analysis. For the third flight (11/03/2016), 3 laps were flown and all were used in the analysis. For the Lloydminster region, 4 laps were flown on 11/04/2016 with 2 used in calculations, and 4 laps were flown on 11/05/2016 with 2 used. One of the discarded laps in Lloydminster on 11/04/2016 was interrupted when the aircraft was buzzed by a CF-18 fighter jet presumably from the base near Cold Lake to the North, which caused the frightened and unamused pilot to deviate from the path.

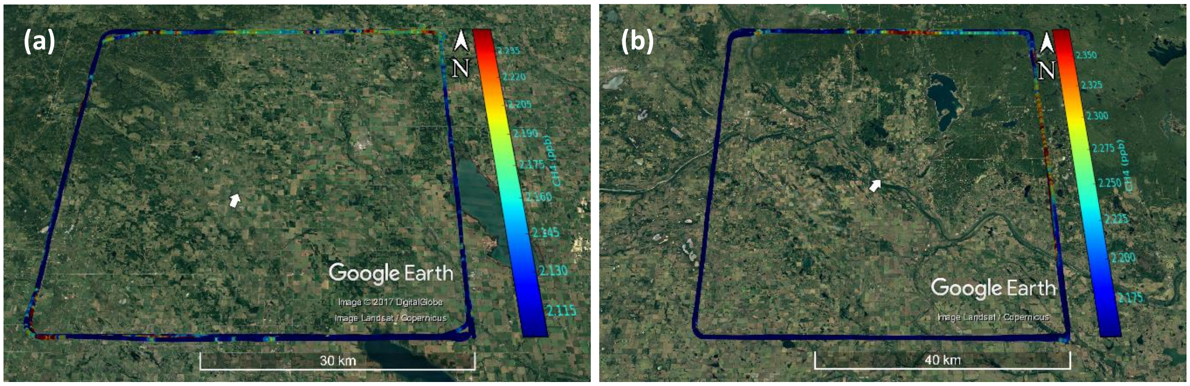

Figure S1: Maps of flights in (a) Red Deer (11/02/2016) and (b) Lloydminster (11/04/2016). General wind direction is indicated by the white arrow at the center of the box. The color scale shows the methane mixing ratio measured during the flight.

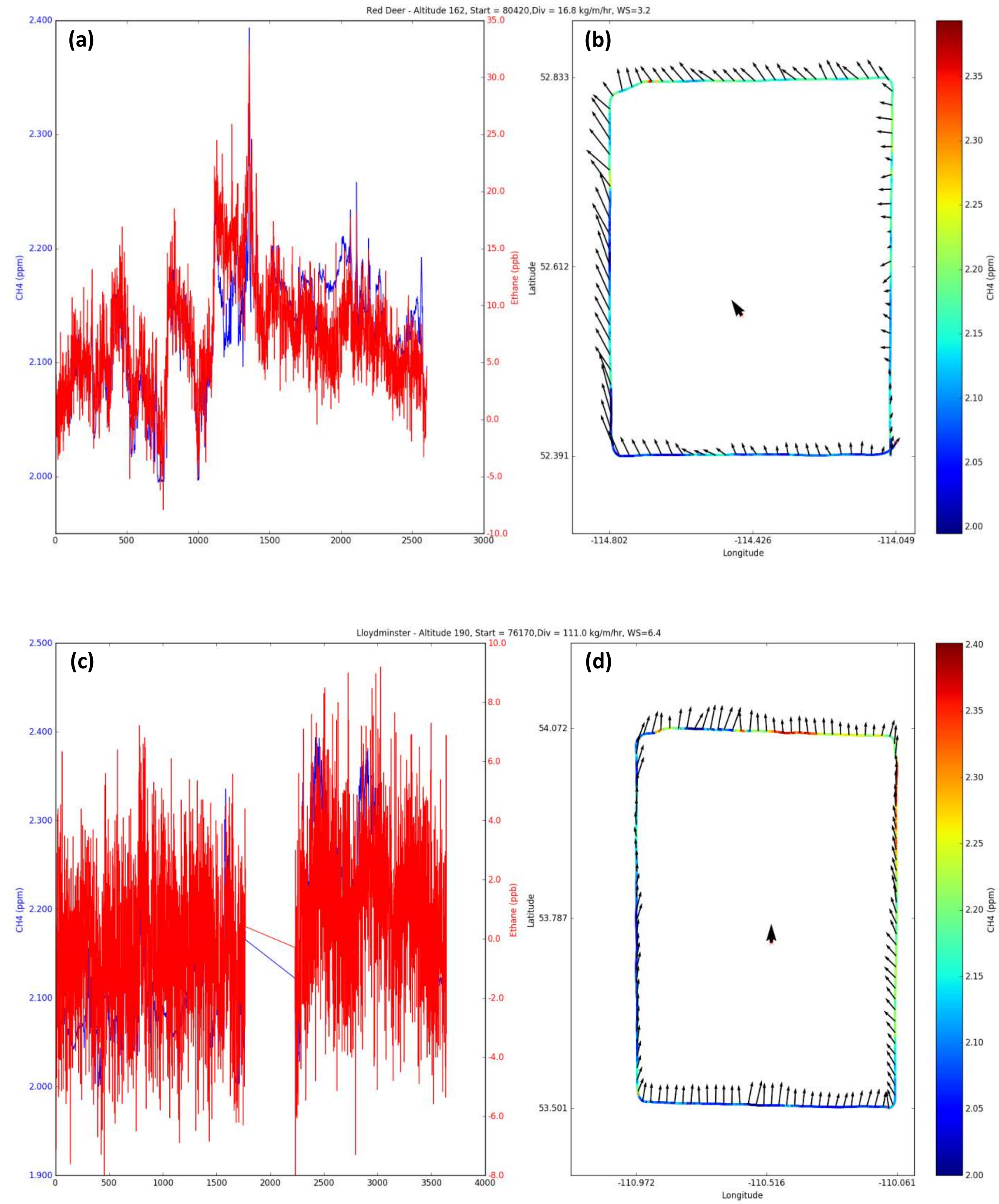

Figure S2: Time series (a, c) for methane (blue) and ethane (red) and the geographic distribution of methane (b, d) for (a, b) one lap flown at 162m AGL over the Red Deer field on 11/02/2016, and (c, d) one lap flown at 160 m AGL over the Lloydminster region on 11/04/2016. The gap in (c) represents the time spent flying a vertical profile of the boundary layer. The geographic plots show the instantaneous wind vector (small arrows around the flight path) as well as the mean wind direction (large black arrow in the center of the flight path).

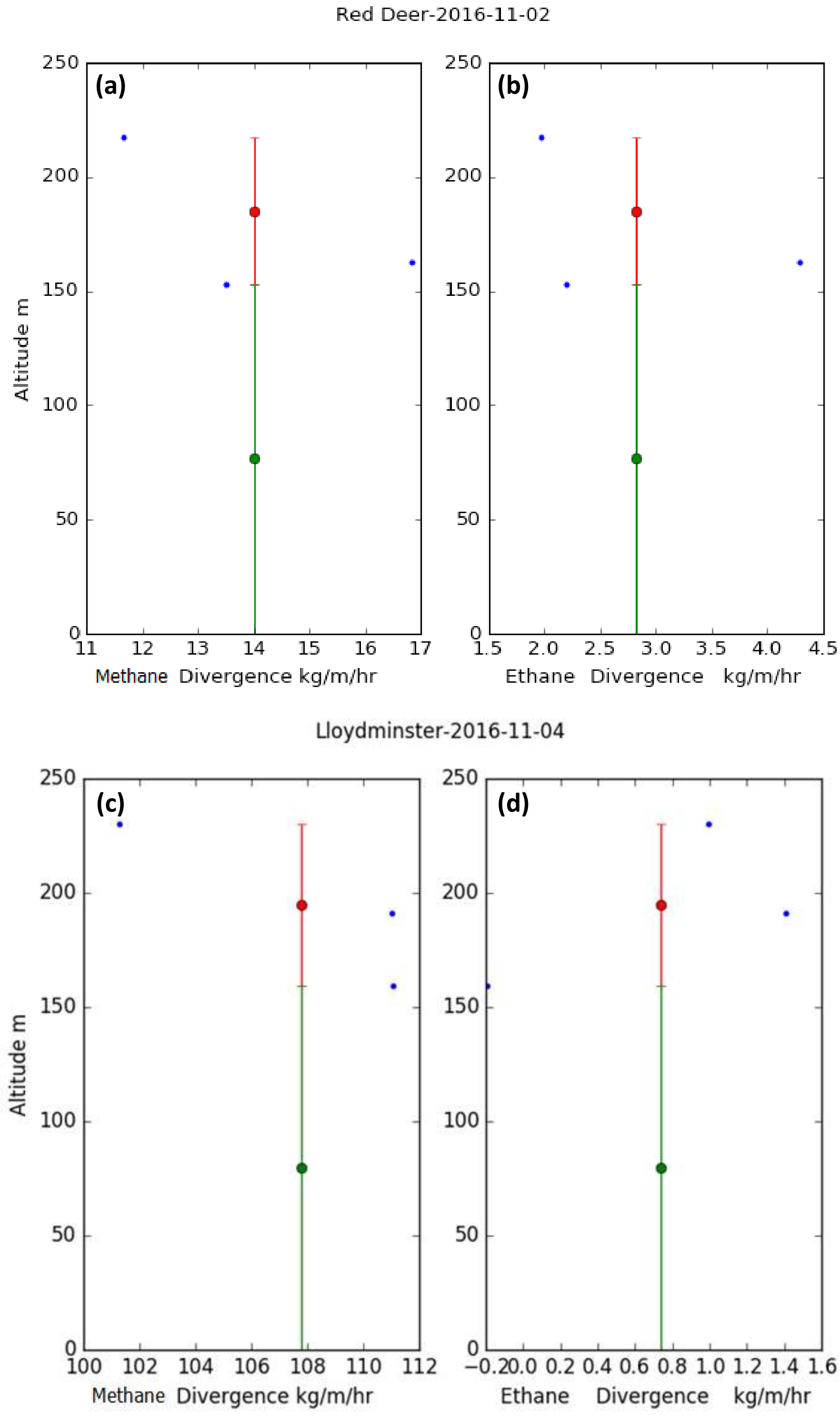

Figure S3: Methane (a, c) and ethane (b, d) flux divergence profiles for the Red Deer (11/02/2016) flight and the Lloydminster (11/04/2016) flight. Blue dots represent individual loop measurements, while the red circles represent the bin average values for altitude intervals represented by the bars. Finally, the green circle represents the estimated flux in the region below our lowest measurement (calculated as the average of the actual measurements).

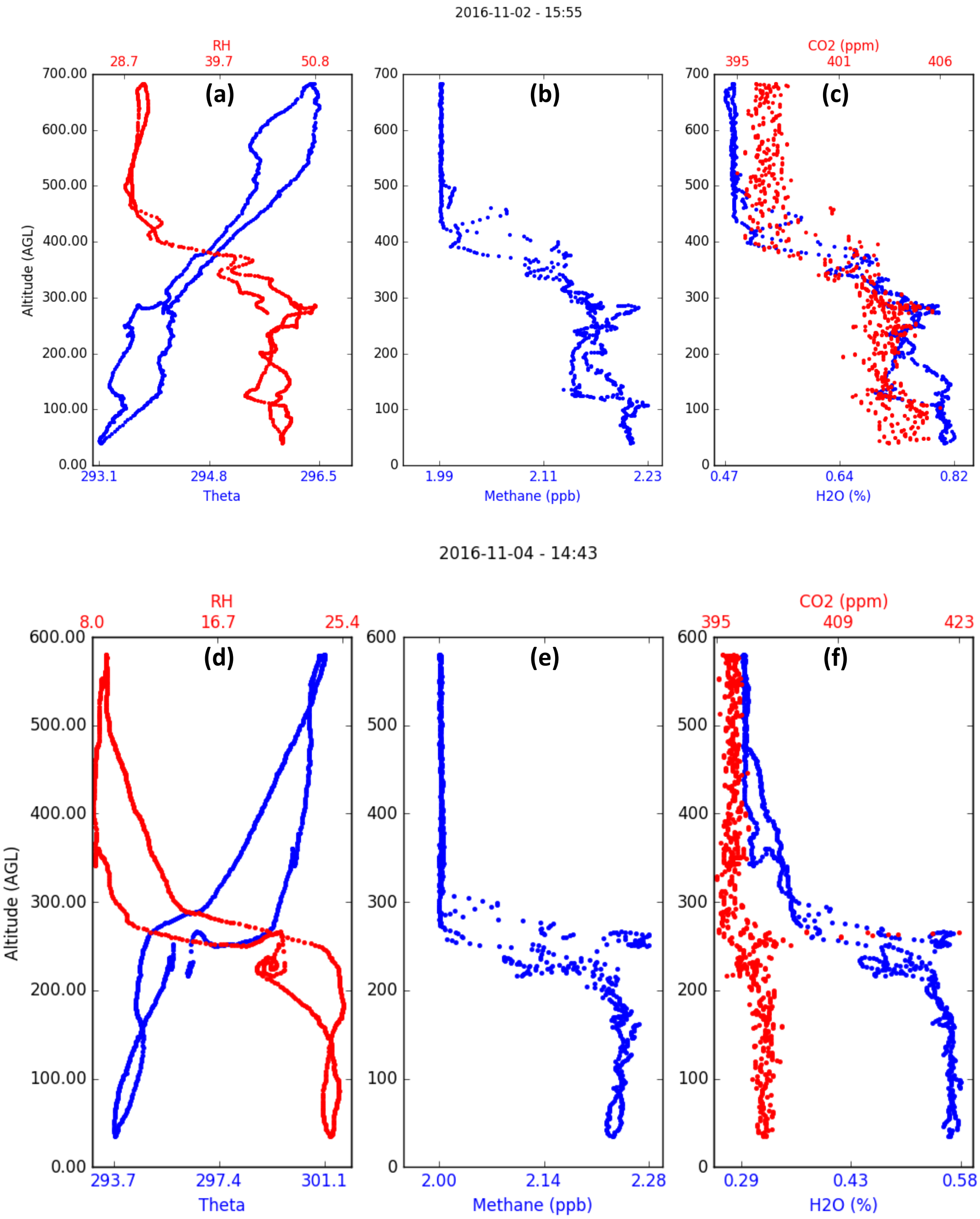

Figure S4: Boundary layer profiles (a, b, c) for Red Deer 11/02/2016 flight and (d, e, f) for the Lloydminster 11/04/2016 flight. These profiles (and many others) were used to track the growth of the boundary layer, evident in the profiles of potential temperature (Theta), relative humidity (RH), methane (CH_4), and water (H_2O).

S.1.2 Lap Selection

The optimal conditions for an airborne measurement are a well mixed boundary that rises above the minimum safe flight altitude, with stable consistent winds. For this campaign, the flights were conducted north of Calgary at a time of year when the sun was near its southern most point (November). The colder temperatures reduced the buoyant motion of the boundary layer and presented a flight challenge as the height of the boundary layer was often below our 150 m minimum flight altitude. Over the course of an individual lap, if part of the lap was above the boundary layer, that entire lap needed to be discarded. This was determined post flight by examining the time series of CH_4, CO_2, C_2H_6, temperature and humidity and looking for times when there was indication of measuring background (above the boundary layer) values.

Figure S5 shows an example of an excluded lap. Each lap included one vertical boundary layer profile, which was excluded from the flux calculation. This vertical profile is labelled at a time index 1429 s, with the arrows pointing toward the start and end of the excluded period. The other label 'Above BL' shows an area where potential temperature ("Theta" on the graph) increased (because the aircraft crossed the potential temperature inversion height) and the methane trace went flat at a value typical of background (1.97 ppm). Each lap was scrutinized in this way for periods when the aircraft was flying above the boundary layer, and, if observed, the lap was discarded.

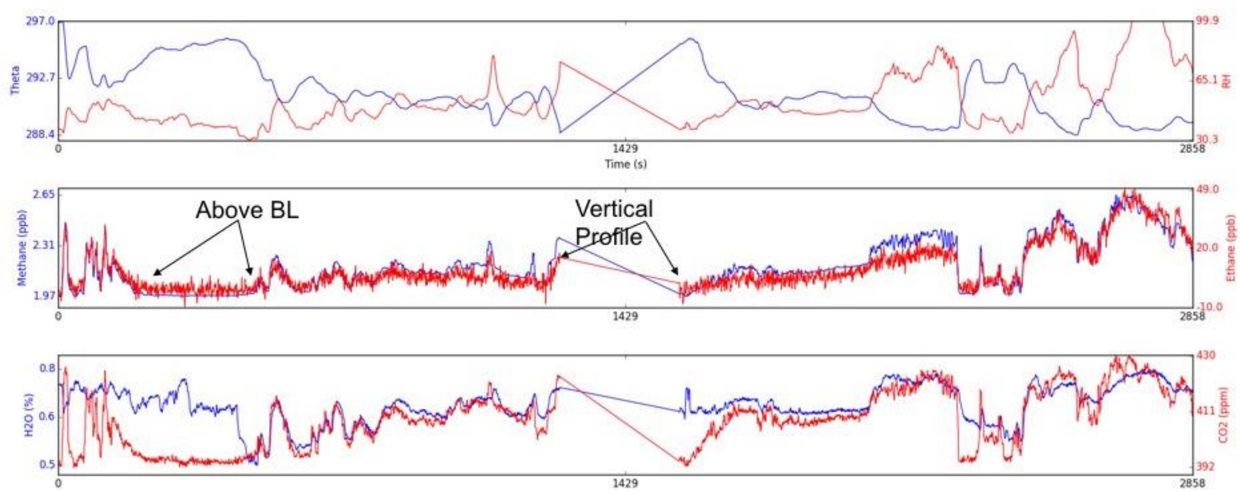

Figure S5: Example data from and excluded lap flown in the Red Deer Region on October 27, 2016.

S.2 Regional Gas Compositions

As outlined in the manuscript, site specific gas compositions were generated using available AER data containing 312,654 useable gas samples associated with 117,206 unique well segments, each identified by a Unique Well Identified (UWI). From these data, estimated compositions at each well or well segment in the province were determined. Finally, using gas production volumes reported at each UWI, distributions of methane and ethane fractions in the produced gas within each measurement region were derived as plotted in Figure S6. In addition, to facilitate best possible estimates of methane emissions from reported whole gas venting volume data, compositions at downstream facilities (e.g. gas plants and gas gathering systems) were determined based on the gas volume weighted average of the compositions of the received gas at that facility.

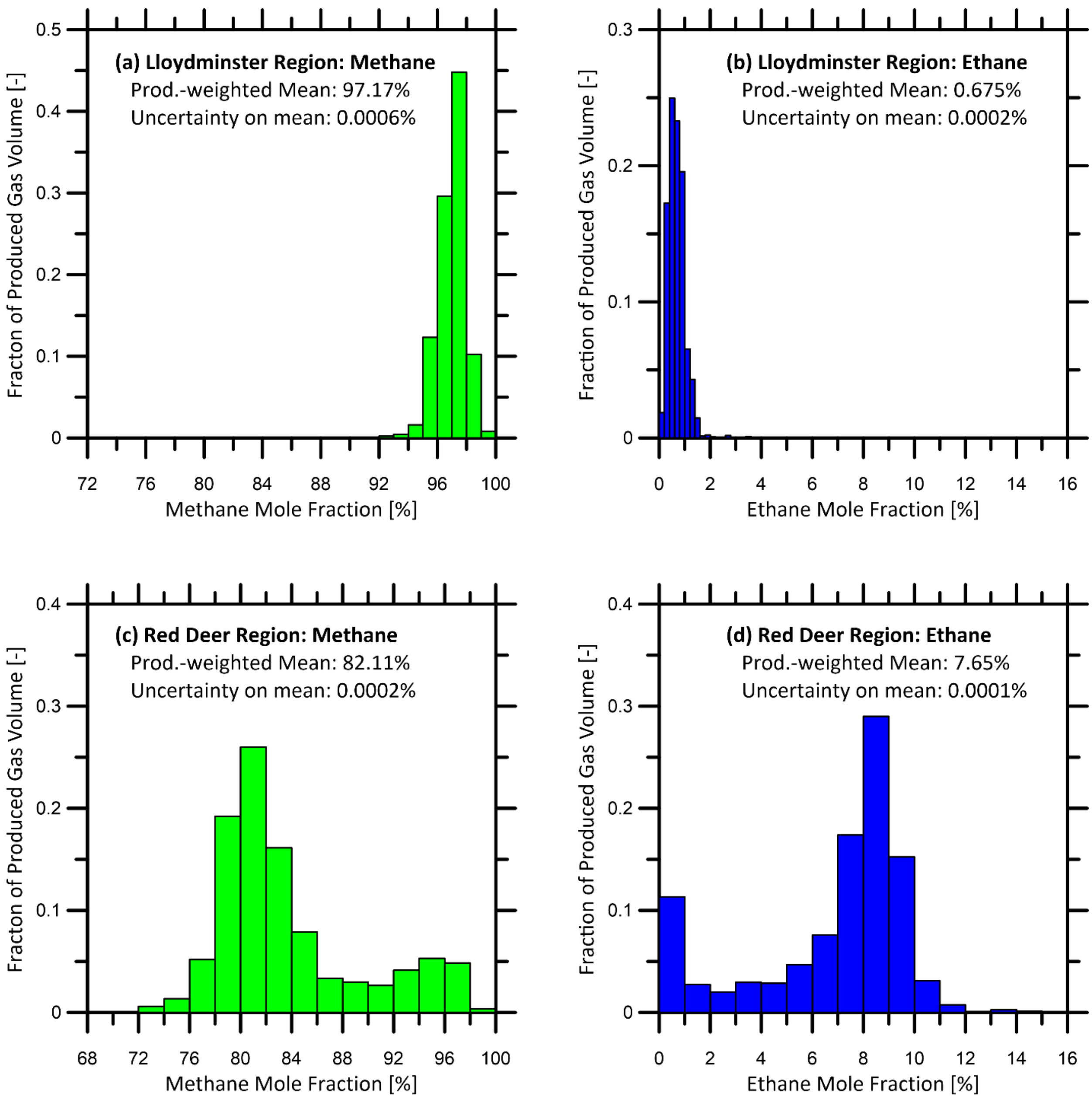

Figure S6: Distributions of Methane and Ethane mole fractions in produced gas volume in the Lloydminster measurement region (a,b) and the Red Deer measurement region (c,d). Gas production weighted mean values are shown on each graph. Uncertainties on these mean values were calculated via a bootstrapping statistical analysis.

Compositions of produced gas in the Lloydminster region were narrowly distributed, with much higher methane fractions, and much lower ethane fractions than those of the Red Deer region. The gas-production-weighted mean methane composition was 97.17% in the Lloydminster region versus 82.11% in Red Deer. Mean ethane fractions were 0.675% in Lloydminster versus 7.65% in Red Deer. Uncertainties on these mean values were estimated using a bootstrapping statistical analysis. For each region, 5000 independent distributions were created by randomly drawing (with replacement) 100 m^3 volumes of gas (resolution of the reported production data) from the distributions in Figure S6, where each bootstrapped distribution for each region represented the same total gas volume. The variation in the means of these bootstrapped distributions is a measure of the uncertainty in the production-weighted mean composition for each region. As reported on the graphs, the very tight uncertainties on the production weighted mean species compositions reflect the very significant produced gas volumes in each region (i.e. large sample size) – 467 million m^3 in the Lloydminster region and 3511 million m^3 in the Red Deer region.

As described in the manuscript the methane-ethane ratio derived from compositional samples in the Red Deer region was used to allocate the biogenic portion of the measured airborne methane flux. Accordingly, the accuracy of the inferred methane flux attributed to oil and gas sources in the Red Deer region is directly influenced by how representative the composition profile in Figure 7(c) and (d), is of the gas emitted to atmosphere in the region. The well activity in the Red Deer region splits roughly into 75% gas sites and 25% oil sites. Typical venting sources in the Red Deer include fuel gas driven pneumatics and compressor vents at both oil and gas sites, as well as, flashing loss from storage tanks at oil sites. In cases where processed fuel gas may be leaked through pneumatics, the methane content could potentially be higher than than the formation composition. Conversely, gas emitted through routine flashing losses from tanks could be expected to have a lower methane content.

Leaks of processed gas, especially on the downstream side of gas plants, are not expected to influence the overall composition profile of the region. For context, within the Red Deer measurement region there are 2053 gas wells and 613 oil wells and 11 gas plants. It is thus more likely that the regional composition profile is driven by well site pneumatics and compressor vents. In a recent report[4], GreenPath Energy Ltd. inventoried pneumatic equipment and associated leaks in a study region that almost completely overlaps the Red Deer measurement region. They estimated an average pneumatic leak rate of 6.54 tCH_4/year per well. Applying this emission factor to the 2666 wells in Table 1 of the manuscript suggests a methane flux of 2.0 tCH_4/h from pneumatic devices alone. Compared to the overall aircraft based estimate of 3.05±1.1 t/h of methane emissions in the region, this affirms the likely dominance of emission sources upstream of gas plants.

Emissions from tanks are similarly not expected to influence the overall composition profile. Routine flashing losses from tanks in Alberta are typically estimated by the operator using site specific gas-oil ratio (m^3 gas/m^3 oil) or gas-in-solution ratio (m^3 gas/m^3 oil/kPa)[5] and reported in aggregate with other reported sources of venting in accordance with Directive 60[6]. Although flashing losses in routine production are generally accepted to be small, if in the extreme, it is assumed that all reported venting from oil sites in the Red Deer measurement region is attributable to routine tank flashing losses, then reported tanks losses would account for roughly 3.6% of the bottom up methane flux estimate. Thus, if tank emissions were driving the overall ethane/methane emission profile of the region, it could only be through fugitive and unreported venting. However, recent airborne studies[7,8] suggest that in areas where tank emissions are large enough to influence the overall emission profile, the underlying cause is likely from abnormal process conditions. This may include undersized or over pressurized separation equipment, as well as, malfunctioning separator dump values and/or control equipment. In each case, these abnormal process conditions allow produced gas to either intermittently, or in some case persistently, vent through the production tanks. This would again result in a composition profile that is consistent with the formation gas.

S.3 References

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