Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-109, 2017
Manuscript under review for journal Atmos. Chem. Phys.
Discussion started: 7 April 2017

Atmospheric Chemistry and Physics Discussions

Mobile measurement of methane emissions from natural gas developments in Northeastern British Columbia, Canada

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Abstract. North American leaders recently committed to reducing methane emissions from the oil and gas sector, but information on current emissions from Canadian unconventional developments is lacking. This study examined the incidence of methane in an area of unconventional natural gas development in northwestern Canada. In August to September 2015 we completed almost 8000 km of vehicle-based survey campaigns on public roads dissecting developments that mainly access the Montney formation in northeastern British Columbia. Six survey routes were repeated 3-6 times and brought us past over 1600 unique well pads and facilities developed by more than 50 different operators. To attribute on-road plumes to infrastructural sources we used gas signatures of residual excess concentrations (anomalies above background) less than 500 m downwind from infrastructural sources. All results represent emissions greater than our minimum detection limit of 0.59 g/s at our average detection distance (319 m). Unlike many other developments in the US for which methane measurements have been reported recently, the methane concentrations we measured at surface were close to normal atmospheric levels, except inside natural gas plumes. Roughly 47% of active wells emitted methane-rich plumes above our minimum detection limit. Abandoned and under-development well sites also emitted methane-rich plumes, but the incidence rate was below that of producing wells. Multiple sites that pre-date the recent unconventional Montney development were found to be emitting, and in general we observed that older infrastructure tended to emit more often (per unit) with comparable severity in terms of measured excess concentrations on-road. We also observed emissions from facilities of various types that were highly repeatable. Emission patterns in this area were best explained by infrastructure age and type. Extrapolating our results across the Montney development, we estimate that the emission sources we located (emitting at a rate > 0.59 g/s) contribute more than 111,800 tonnes of methane annually to the atmosphere. This value exceeds reported bottom-up estimates of 78,000 tonnes for all oil and gas sector sources in British Columbia, of which the Montney represents about 55% of production. The results also demonstrate that mobile surveys could be used to exhaustively screen developments for super-emitters, because without our intensive 6-fold replication we could have used single-pass sampling to screen 80% of Montney-related infrastructure. This is the first bottom-up study of fugitive emissions in the Canadian energy sector, and these results can be used to inform policy development in an era of methane emission reduction efforts.

1 Introduction

As global energy needs continue to rise, oil and gas operators are increasingly recovering natural gas from less-permeable reservoirs, such as tight sandstone and shale, despite environmental concerns surrounding extraction methods. Unconventional techniques, such as horizontal drilling and multi-stage hydraulic fracturing, can be used to stimulate production of natural gas directly from the source-rock in a petroleum system, ultimately increasing the total quantity of marketable natural gas. Presently, Canada is the fifth-largest producer of natural gas worldwide, with enough unrecovered natural gas to sustain 2013 national consumption levels for 300 years (NEB, 2016). More than 68% of Canada's remaining 1087 trillion cubic feet of marketable natural gas reserves is in unconventional reservoirs (NEB, 2016). By 2035, Canadian natural gas production is predicted to increase 25% above 2013 levels, and this projected growth is largely attributed to unconventional methods of extraction such as horizontal drilling and multi-stage hydraulic fracturing.

Compared to coal, many consider natural gas to be a preferable fossil fuel because it emits 50-60% less carbon dioxide (CO_2) during combustion (NETL, 2010). Natural gas has consequently been deemed a transition fuel because it allows for continued fossil fuel exploitation with ostensibly less environmental impact. However, the primary component of natural gas is methane (CH_4), a very potent greenhouse gas (GHG). Over a 100-year timespan CH_4 has a radiative forcing greater than 30 times that of CO_2 (IPCC, 2014). A recent study suggests that if more than 3.2% of total natural gas production is emitted into the atmosphere during upstream operations, the environmental benefits of natural gas, compared to coal or oil, are negated (Alvarez et al., 2012). Therefore, to comprehensively analyze the GHG footprint of different fuel types, it is necessary to consider industrial emissions during upstream operations; these include both vented (intended) and fugitive (unintended) emissions from wells, facilities, and pipelines, during extraction, production, and processing.

"Well-to-wheel" life-cycle assessments (LCA) are a method of comparing the environmental impact of fossil fuels in relation to their carbon emissions. This type of LCA sums all estimated carbon outputs, including emissions during upstream operations, transportation, and combustion. Several recent LCAs suggest that the carbon footprints of unconventional natural gas developments exceed those of conventional natural gas developments (primarily due to emissions during well completions), but that coal developments have the worst overall emissions impact (Hultman et al., 2011; Jiang et al., 2011; Skone, 2011; Stephenson et al., 2011). Contrastingly, another study suggests that conventional natural gas has a slightly higher carbon footprint than unconventional natural gas because of emissions during the liquid unloading process, but that coal remains the fossil fuel with the highest life-cycle carbon emissions (Burnham et al., 2012). A controversial study by Howarth et al. (2011) concluded that a large amount of atmospheric emissions associated with upstream shale gas operations render its environmental impact more severe than coal. This study has been widely disputed for overestimating CH_4 emissions during upstream shale gas processes by not acknowledging that gases emitted during well completions are often flared or controlled by performing reduced emission completions (CNGI et al., 2012). The variability of results from these recent "well-to-wheel" LCAs demonstrates that total upstream emission volumes are difficult to quantify using estimated values of emission incidence. It is important to know what percentage of infrastructure in a development is actually emitting, and active detection and measuring techniques are required to gain this understanding. It is also necessary to acknowledge that emission incidence could vary drastically between

Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-109, 2017
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developments for many reasons including operator practice, and the properties of the geological formation that the gas is being extracted from.

The common infrastructural sources of fugitive emissions are poorly understood, particularly in unconventional natural gas developments where the extraction practices are newly implemented. Detection of atmospheric fugitive emissions from upstream sources has previously been attempted with top-down methods and specific ground-based techniques. Top-down measurements involve airborne (Karion et al., 2013; Caulton et al., 2014), and remote sensing (Govindan et al., 2011; Schneising et al., 2014) measurements. These methods often cover large areas in low resolution proving difficult to identify exact sources of emissions. Ground-based techniques, including infrared camera leak inspections (Mitchell et al., 2015), well injection tracers (Mayer et al., 2013), and soil gas sampling (Beaubien et al., 2011; Romanak et al., 2012), are often too labour intensive to be convenient for use in large oil and gas developments.

Although a recent study assumes that around 63% of infrastructure is emitting in the Barnett Shale (Rella et al., 2015), the majority of inventory studies do not report the incidence of emitting and non-emitting infrastructure. Ultimately, CH_4 management will entail a coordinated targeting of super-emitters, and reduction of overall emission incidence. So, studies that build geospatially distributed information on emission incidence in large populations of infrastructure is a logical next step, because it is the best means of identifying trends across vast developments, behavioural patterns of operators, and the impact of infrastructure age on emission frequency and severity. Mobile screening methods similar to EPA OTM33A (Brantley et al., 2014), even that simply detect incidence, are extremely valuable because emission factors are already available and can be applied uniquely to known emitters so that volumes can be estimated to a reliable degree.

In this study we used a multi-gas (CO_2, CH_4) mobile surveying method that uses ratio-based gas concentration techniques and wind data to detect and attribute on-road CH_4-rich plumes to the infrastructural sources of natural gas developments in northeastern British Columbia, Canada. Our study area is commonly referred to as the Montney, in reference to the extensive, petroleum-rich, geologic formation covering 130,000 km^2 aerially between British Columbia and Alberta (BC Oil and Gas Commission, 2013). It was first recognized as an unconventional petroleum reservoir in 2007, and attempts at accessing its resources were accomplished with horizontal drilling and multi-stage hydraulic fracturing. These unconventional methods yielded 4-5 times more natural gas from the Montney formation than conventional techniques that were attempted prior to 2007 (BC Oil and Gas Commission, 2012). Since then, unconventional production has increased significantly, with thousands of new wells being drilled in northeastern British Columbia.

While the Montney is a challenging first target for mobile emissions surveying because of its spatial extent and lack of accessibility (many poor condition roads), it is a sensible first choice given that its emissions have not been measured independently of industry and government, and because the production mode is largely unconventional - and therefore subject to a higher degree of scrutiny. The less-permeable, natural-gas hosting portion of the Montney formation is located in BC, a province that has generally been very progressive on many issues of environmental stewardship, so there is a broad interest in emissions quantification and environmental performance.

2 Methods

2.1 Field Measurements

Between August 14 2015 and September 05 2015 we collected atmospheric gas concentration data along six pre-planned routes in the Montney formation of northeastern BC (Fig. 1). We designed the routes to come as close as possible to a high number of hydraulically fractured natural gas wells and their associated processing facilities, while also incorporating a variety of operators and infrastructure age profiles. These were on-road campaigns only, and did not approach well pad infrastructure or facilities.

In total we surveyed 7965 km at 1 Hz frequency and the average route length was 248 km. Four of the six routes dissected natural gas developments containing hydraulically fractured wells. One route targeted an older development in the same area that mainly produces oil; this route was intended for preliminary comparison between conventional oil and unconventional natural gas developments. The sixth route was located outside the perimeter of concentrated natural gas infrastructure, and was intended to act as a control. We surveyed four of the routes six times throughout the field campaign, and the two remaining routes (including the Control Route) three times each. Repetitions of surveys were necessary in part because wind directions could not always be guaranteed favourable (truck downwind of wells), but also so that we could obtain statistics on emission persistence, and confidence that we were indeed targeting CH_4-enriched plumes originating from natural gas infrastructure. Repetitions of each survey route included both morning and afternoon drives to incorporate varying atmospheric conditions.

The mobile surveying platform used to collect these data consisted of an LGR Ultraportable Greenhouse Gas Analyzer (Los Gatos Research Inc, San Jose, CA, USA) Off-Axis Integrated Cavity Output Spectrometer (1σ instrumental errors of <2 ppb at 1 sec), to measure raw atmospheric concentrations of CO_2, CH_4, and H_2O. A high volume (7 lpm) air pump brought air to the analyzer from the front of the vehicle through 6 mm ID tubing. During post-processing we applied corrections for lag times between the intake filter and the gas analyzers. An NM 150 weather station (New Mountain Innovations, Old Lyme, CT, USA) was located 1.5 meters above the vehicle to collect wind and weather data (with instrumental errors of $\pm1.5°$ for wind direction and $\pm4\%$ for wind speed). Gas species concentrations and wind velocity measurements were collected every second while driving. Wind velocity measurements were corrected for both the direction and speed of the vehicle, and we geo-located all data-points using an onboard GPS.

2.2 Identification of Natural Gas Emissions

Both CO_2 and CH_4 exist, and vary, naturally in the atmosphere. We had to account for this variance in order to identify anomalous measurements that were potentially sourced from natural gas developments. Additionally, only infrastructure upwind of our truck could be sampled, so we used both geochemical and geospatial methods of attribution.

To accommodate the fluctuating background concentrations of CO_2 and CH_4, the traditional approach would either be for the user to set a concentration threshold above which a reading would be considered an anomaly, or for a dataset minimum value to be used as the background (as in Hurry et al. (2015)). However, our surveys were normally multiple hours long and brought us through various land use types, making a pre-set threshold or global minimum approach unusable. Instead, we used

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a simple iterative deconvolution method in which we reset the ambient "background" concentration of each gas at a specified time interval, called the Running Minimum Reset Interval (RMRI), and where we iteratively scaled the RMRI until we had maximized the number of (consecutive multi-point) above-background ("*excess*") ratio emission anomalies. In other words, an optimal RMRI was determined for each survey by iteratively applying a suite of RMRI values (60s to 1800s, at an interval of 60s) to our datasets, subtracting the background, and evaluating the number of multipoint $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ < 150 excursions. As RMRIs shortened, more small emission anomalies were exposed, by about 2-3 times relative to the dataset minimum approach used by Hurry et al. (2015). However, when the iteration approaches very small RMRIs (<180 s), it consistently caused the total number of anomalies to increase (often by a factor of 10), in particular for anomalies of extremely small concentration. This was expected because when concentration resetting is done too quickly, it overlaps in the temporal domain with instrument and other random noise, causing every departure to seem anomalous relative to the recently reset background. Our optimal RMRI was taken to be the point at which anomalies were maximized, but also where we avoided the rapid noise-associated increase associated with extremely short RMRIs. We applied this method separately to each of the 30 surveys. RMRIs of about 300 s were normally most favourable for the resolution of $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ < 150 excursions, but for some surveys in more consistent terrain (or weather) longer RMRIs proved better. This means that for most surveys, the background concentration for each gas was reset every ∼300 s, to the lowest recorded concentration value during the preceding 300 s. While this background subtraction technique improves the resolution of localized plumes, it should be clear that it impedes the resolution of larger regional anomaly features, or mega-plumes, because they may in fact form an artificially elevated background that persists across the 300 s scale. Combustion values were also recorded along the routes when $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ exceeded 1000, and were related to vehicle tail-pipe emissions and industry.

Methane-rich plumes were identified in areas of super-ambient $\mathrm{CH_4}$ concentration (after background subtraction), in which successive datapoints of depressed $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ ratios were located. The $\mathrm{CO_2}$:$\mathrm{CH_4}$ ratio of normal air is roughly 215, and $\mathrm{CH_4}$-rich plumes from natural gas sources are substantially more depressed at the point of origin (the Montney does contain some $\mathrm{CO_2}$ in variable, but generally super-ambient, concentrations). We used ratios of these gases in detection instead of raw $\mathrm{CH_4}$ concentrations, because ratios are more conservative than concentrations in valleys and other areas where pooling of gases is common, and fewer false positives are likely. Since fugitive and vented gas sources might be highly diluted in air, their presence will not significantly affect the normal bulk ratio. In this case, the $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ ratio will record the anomalies with a higher degree of fidelity. This excess $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ approach has been used as a fingerprinting tool in oil and gas environments by Hurry et al. (2015). Here, we assumed that $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ ratios < 150 were representative of significant departures from the normal natural ratio, and potentially indicative of an exogenous $\mathrm{CH_4}$ source locally. In order for a natural gas related plume to be identified, we had to detect > 3 successive datapoints with $e\mathrm{CO_2}$:$e\mathrm{CH_4}$ ratios < 150.

2.3 Emission Source Attribution

Publicly-available files from the BC Oil and Gas Commission (BC OGC) (acquired July, 2015) of all oil and gas infrastructure in the province were used to perform geospatial analysis. We modified these files to exclude temporary or virtual facilities, such as those in place only during well drilling, or artificial facility entries used to record regulatory information. Otherwise,

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all in-place oil and gas infrastructure both upwind, and within 500 m of an on-road CH_4-enriched anomaly, was considered a possible emission source. Our database included active, developmental, and suspended well pads, and various facility types. When we detected $e\mathrm{CO}_2$:$e\mathrm{CH}_4$ < 150 excursions on-road, and infrastructure was present upwind, and also within the target radius of 500 m, the infrastructure was flagged as a probable emission source. All observations were databased in SQL format databases, with processing, statistics, and plots done using R (R Core Team, 2016).

No unique thermogenic tracer was used in this package of surveys to discriminate biogenic CH_4 sources, such as cattle that may have been present on the well sites at the time of surveying. However, repeated surveying of each route increased our confidence that we were tagging stationary natural gas infrastructural sources. Persistence is also an important metric not only for detection, but because many of these fugitive and vented emissions are episodic in nature. Though the infrastructure is stationary, the emissions are not necessarily continuous, and gas migrations, surface casing vent flows, leaks, and tank vents, are all known to have a temporal component. Additionally, maintenance activities may have been occurring onsite at the time of survey, which would generate a non-persistent emission pattern and occasionally we were proximal to drilling or fracturing operations.

3 Results and Discussion

In total, we performed 30 surveys between 200 - 550 km in length each. We surveyed infrastructure managed by more than 50 operators along six different routes. Compared to some oil developments in western Canada, natural gas developments in northeastern British Columbia are spread out, and therefore required a considerable amount of driving. It was not possible to secure a Control Route that was totally free of oil and gas infrastructure, but our Control route did have a density of infrastructure that was much lower than that of other routes, with intervals that were relatively unpopulated.

3.1 Measured Gas Signatures

Methane was the gas of primary interest for this study, and bulk CH_4 values were in general not appreciably different from background air. Mean CH_4 for the study was 1.897 ppm with σ=0.084 ppm (n=444515). Max and min were 8.148, and 1.819, respectively. Since the background was very stable, anomalies detected near oil and gas infrastructure were both obvious, and short-lived. These bulk concentrations contrast with those measured for other developments. For example a study in the Barnett Shale measured a mean CH_4 concentration of 11.99 ppm, with a median of 2.7 ppm, in residential fringes surrounding shale gas development (Rich et al., 2014). The Barnett Shale has about three times as much infrastructure in half the area, but the mean departures in the Barnett exceed the maximum departure in this study. In the Montney, ambient CH_4 concentrations were seldom measurably different than global norms (about 1.850 ppm but regionally dependent). As a result of the stable background, combined with the deconvolution approach, we were able to detect the presence of emissions hundreds of metres away from infrastructure using the mobile survey method. On average, most of our detections were at a mean distance of 319 m from the infrastructure we were sampling (Figure 2), and natural gas emissions were detected at a mean distance of 314 m from the probable emission source.

Figure 3 shows the aggregate (all survey repetitions) kernel density plots of eCO_2:eCH_4 for the survey routes (ratios of CO_2 to CH_4 above ambient). In each, we see a peak of signatures near ∼215 which is representative of natural. Though most of the natural should be filtered out in background subtraction, some of the background signature remains in our datasets during the initial increase and decrease in CH_4-enriched peaks. The kernel density plots show that, in all of the survey routes except the Control, we see a population of relatively CH_4-enriched anomalies to the left of natural (numerically low eCO_2:eCH_4) that are the result of localized plumes from natural gas development. The Control Route lacked a population of enriched CH_4 values, which was expected because the density of infrastructure was comparatively low.

We used the gas concentrations collected on all three surveys of the Control Route to calculate our probability of falsely detecting a CH_4-enriched plume. To do this, we calculated the percentage of datapoints more than 5 km away from any oil or natural gas infrastructure that our method falsely interpreted to be part of a plume. Using this method we calculated our probability of a false plume detection to be 0.2% on our Control Route. It should be noted that the Control Route did have other types of industry (such as a pulp mill and active logging) which were not present on the other routes. Therefore, this confidence in plume detection is a conservative calculation that can be applied to all five other routes that we surveyed as a part of this study.

3.2 Emission Sources and Trends

Once geochemical plume detection was complete, existent nearby oil and gas infrastructure was tagged if it was a probable emission source. An example of this binary result is presented visually in Figure 4, where infrastructure is shown in red when tagged as emitting, or in green when emissions were absent. However, we rarely dealt with the maps directly because our aim was to investigate industry-wide patterns, and drivers, across types and age classes of infrastructure and operators. For further analysis, these binary data were folded into datasets along with infrastructural characteristics extracted from the geospatial databases. Like geochemical attribution, there is some potential of false positives when geospatially attributing plumes that we observed on-road. While control routes allow us to be very confident about the existence of plumes, we can be somewhat less certain about their precise origin. In areas of low infrastructural density, geospatial attribution confidence is maximized. But in areas of high density, it is possible that emissions from a suspected source are actually being emitted from a co-located battery, gathering pipeline, or other. A FLIR camera or other would be required to trace each plume precisely to the source gasket, vent, or soil area, and that work was beyond the scope of this study. Therefore, the following section should be considered as an analysis of probable emitting infrastructure, *plus* possibly emitting co-located associate infrastructure.

Of the various types of infrastructure that we passed, well pads were the most commonly occurring. Several classes of wells were listed in the geospatial databases, including active, abandoned, canceled, completed, and under development. In this study, emission persistence was defined as the number of passes on which a CH_4-rich plume was attributed to a piece of infrastructure, divided by the number of times we surveyed that infrastructure in the downwind direction. In order for a piece of infrastructure to be classified as an emission source, it had to have > 50% emission persistence. Where our technique is tuned to resolve small localized emission plumes, we expect that even for continuously emitting infrastructure, we might not detect any emission on each pass when detection should theoretically be possible. Of course, atmospheric conditions have a significant impact on the downwind detectability of emissions. In buoyant and unstable atmospheres, emission plumes will have a tendency to rise, and may not be detected reliably on the ground at distances of several hundreds of metres. As such, we would expect that the probability of detecting emissions on 100% of passes is lower than the probability of detecting emissions on 50% of passes. However, even a figure of 50% persistence (normally detected 2-3 times) indicates that there is high likelihood of a continuous emission at the site, though it might be of small scale which is why we detect it only episodically. In general, fugitive emission studies in the literature are not well replicated, but replication helps both build confidence in detection, and a story around the themes of severity and persistence. Operators and policymakers may find value in these data when prioritizing sites for further investigation, or mitigation.

Figure 5 presents the fractional emissions (emitting/surveyed) for each class of wells, presented in a regression form. Each point shows the aggregate emission frequency score for each of the six routes, only for emissions that were detected on more than half the transits past the well (50% persistence), and when the wind was in a favourable direction for detection. In most cases, that means that we were able to replicate the detection at least twice, if not up to six times. And, as it was unlikely to have cattle permanently stationed underneath or around the infrastructure, the replication gave us confidence that we were in fact predominantly detecting anomalies sourced from stationary oil and gas infrastructure. We surveyed more Active wells than any other type, and their emission frequency was highest. Some level of repeatable emission was associated with almost half (47.7%) of the active wells. As can be seen from the right-hand column in Figure 5, this 47.7% fraction of emitting Active Wells was detected not only at 50% persistence (our base criteria) but also at 100% persistence - meaning that each emitting Active well was detected on each of up to six passes on different days, and under different meteorological conditions, regardless of size. The 50% and 100% trends in Figure 5 mirror one another very well, and are probably capturing the same signal.

Aside from Active wells, Figure 5 shows other well types that were also identified as probably emission sources. We calculated an emission frequency of 26% for Abandoned, 25% for Cancelled, 30% for Completed, and 27% for the class defined in the databases as Well Authorization Granted, most of which were somewhere in the stages of development during our visits. Of these well classes, the Abandoned wells were the largest in number after the Active wells on our surveys. For these categories, emissions were detected on a more episodic basis than for active wells, by comparing the lefthand, and righthand, columns which represent 50% persistence, and 100% persistence, respectively (Fig. 5). For the 100% persistence columns, the number of pieces of infrastructure emitting was lower, and generally the incidence (slope) also decreased slightly.

While the frequency of emissions from well pads tended to be high, the concentration severity tended to be low. As noted earlier, no concentration above 8.148 ppm was recorded during the surveys themselves. Most of the anomalies were small-scale, and were detected at roadside as CH_4 excursions on the order of $\sim$0.1 ppm. Regardless of the route on which the anomalies were detected, all infrastructure had roughly the same frequency of emission, because all points tended to fall along the same regression line (Fig. 5). While there might be appreciable inter-operator variability at the small scale, these sorts of statistics are expected because emissions are related to the type of infrastructure that sits in service, post-fracturing. This infrastructure is of course similar across the entire development, so it should not be surprising that well pads tapping the same formation 100 or 200 km apart might still have similar emission frequencies when the infrastructure of many operators are statistically bundled together. At the large scale, emission frequency might be an inherent property of the development, related to fluid type and handling, needed infrastructure, accessibility, and operator best practice.

Of the Active wells, a portion were defined by the OGC databases as Production wells, and another portion as Undefined. Only Active Production wells were predictable emitters, with high statistical coherence from route to route (Fig. 6). We expect that some of the wells in the Undefined category were under development, in production, or could have been recently abandoned. Some in the Undefined category may also have even been active water disposal wells, from which we would not generally expect emissions. At some points during this study, the accuracy of available infrastructure data was questioned, because interpretation of trends depended in large part on accurate infrastructure inventories. Maintaining such inventories would be a challenge for regulators, particularly during periods of fast growth, because their capacity to update these publicly-available datasets might become temporarily overwhelmed. In many parts of Canada similar databases are often years out of date, which would present a possible barrier to studies similar to this one, where industry trends are explored using ground-based surveying in conjunction with public datasets.

The number of facilities sampled was overall much lower than for well pads, which simply relates to the relative distribution of facilities as compared to well pads. With all facility types included, we found 32% of surveyed facilities to be emitting (Fig. 7). Compressor stations appear to emit the most frequently (slope = 0.7 or 70% incidence), which would be expected based on the results of Omara et al. (2016) and others. However, a number of zero-detects anchor the lower part of the Compressor stations curve, so the overall relationship is not significant. More compressor stations would need to be sampled in order to arrive at a statistically significant estimate. Also, these larger compressor facilities may emit from a height significantly higher above ground level than normal well pad infrastructure, which makes emission frequency measurements less reliable, and certainly conservative. In other developments where the road network allows for fuller transits around such stations at increasing distances, mobile surveying might be a good approach. But in the Montney, accessibility is often limited. Shared Facilities, Compressor Dehydrators, and Satellite Batteries are more easily targeted, however, and we observed persistent emissions (50% level and 100% level) from all types at a frequency between 11% and 28%. Statistical probabilities (p-values in Figure 7) suggest that individual Batteries did not emit measurably.

Figures 6 and 7 present only anomalies that were repeated on more than 50% or 100% of the passes when we were within the target radius of the infrastructure, and downwind. Figure 8 shows the full spectrum of Occurrence (n) vs Persistence (%) of the emissions across repeat surveys. In the top left hand panel, it is clear that a group of about 60 Active wells emitted persistently, 100% of the times they were surveyed. In some cases, we detected these emissions on all six survey repeats on different days, and under different weather conditions. As discussed earlier, it was predominantly the Active wells that emitted at 100% persistence, though several Abandoned and Canceled wells were also highly persistent emitters. Emissions from the Undefined wells were detected on a more episodic basis. For fluid type, it was generally Gas wells that were the most persistent emitters. Oil wells were tagged as emitters more episodically. The majority of facilities emitted at 50% persistence, although no facility type dominated this trend. As can be seen from Figure 8, there is also an abundance of infrastructure that emitted at the 25% persistence level. As one moves to the right along the x-axis in Figure 8, emissions are more certain, less episodic, and likely also larger in magnitude - enabling more frequent detection across all atmospheric conditions.

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Infrastructure type is a potential driver of emission patterns, and Allen et al. (2013) has, for example, identified a large discrepancy between valves used in different regions of the US, with some being less emission prone. We did not have data on specific infrastructure pieces, but we did have information on ownership and operator size (via number of sampled pieces of infrastructure), and also age. Our results show that newer infrastructure in the Montney emits less frequently, but frequency increases for older infrastructure (Fig. 9). However, since most of the infrastructure is new (large dots on Figure 8), relatively little of it emits - presumably because of improved modern practice, integrity, and better design of new valves, seals, and flange gaskets etc. There was a package of old infrastructure (> 50 years) in the Montney emitting with 100% frequency. While the older infrastructure emitted less often on our surveys, these select pieces are reliable emitters. Infrastructure from larger operators tended to be somewhat lower, but this trend is anchored by a small number of small operators with 100% emission frequency at both 50% and 100% persistence. It is important to note that many large operators grow through acquisition of infrastructure that previously belonged to smaller operators. As a consequence they will often inherit the environmental performance of companies whose assets they buy, and it may take some time to bring these sites in line with company expectations, which will skew our interpretations here.

The bottom two plots in Figure 9 show severity of emissions (as measured by $e\mathrm{CH_4}$ at roadside within the anomalies) as a function of well age and operator size. These concentrations are shown "as recorded" and have not been corrected for dilution within the instrument cavity, and are therefore lower than they would have actually been if we were not in motion but stationary within the plume. However, these figures still provide a useful relative index. Overall, we see that the older infrastructure (> 50 years) has slightly elevated emission severity on the road. We did not note any clear relationship between emission severity and operator size.

There is not a geographic trend to the emissions we detected in the Montney; however, it is clear that certain areas, and potentially their associated infrastructure and practices, result in a higher number of emitting pieces of infrastructure (Fig. 10). Since many studies have shown that super-emitting sites are generally responsible for over half of all observed emissions, a realistic first-order reduction strategy would be to focus on super-emitter Leak Detection and Repair (commonly referred to as LDAR). This focuses attention on the problematic infrastructure and operators, and does not share the cost burden across companies that have already invested heavily in emission reduction technology and leading best practice. It is feasible to detect super-emitters through exhaustive survey campaigns, even from roadside campaigns such as this one. Super-emitting sites, so long as they are near ground level, should be obvious from a single survey, and the logical follow-up would be to verify emissions sites with Forward Looking Infrared Cameras (FLIR) and other techniques. The preferable strategy would, however, be to conduct full loops around each well pad exhaustively, because that would be a much better index of emissions where the surveyor can make a certain and informed judgement based on one pass, because the survey was not dependent on wind direction.

3.3 Minimum Detection Limit Analysis

Minimum Detection Limits (MDLs) allow emission detection studies to identify the measuring capabilities of the method being used, and also to understand the minimum emission inventory within a development. Direct source measurement techniques

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Manuscript under review for journal Atmos. Chem. Phys.
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often have lower MDLs than remote survey studies because the measurements are taken at the emission source over a longer period of time and often within a closed bag. For example, a study by Allen et al. (2013), that detected well pad emissions onsite, had an MDL of < 0.001 g/s. Not surprisingly, MDLs for truck-based surveys are lower, as noted in Brantley et al. (2014). In that study, they came within an average distance of 57 m of the emission sources and collected data for 10-20 minutes at each site of >0.1 ppm CH_4. This translated to a MDL of approximately 0.01 g/s. In comparison, we were detecting emissions from farther away (319 m on average), and recorded gas concentration data for < 20 seconds. However, our method of background subtraction and ratio-based plume identification allowed us to detect smaller concentration anomalies with confidence. Since concentrations will decrease exponentially away from a release source, small concentrations detected at distance could still represent moderately large emission severity. In order to estimate MDLs for this study, we established MDLs for various detection distances using cavity dilution experiments, followed by dispersion modelling.

Dilution in the instrument's measurement cavity is a function of anomaly duration (plume width, plus transit speed across plumes), and cavity size relative to pump rate. In a laboratory experiment we simulated dilution within the instrument using short injection pulses across a wide range of field conditions. We found that for realistic field conditions, the mean level of dilution was about 70%. In other words, the short pulses resulted in only 30% of the potential concentration deviation. Or, that observed concentrations were on average of 3.3 times lower than the actual ambient concentration that would be observed by a stationary analyzer. This dilution factor must be considered when interpreting our concentration readings at road-side, and also while calculating emission volume estimates. While it would possible to estimate a MDL for the hundreds of plumes separately, for simplicity we chose instead to focus here on mean MDLs.

Following the dilution experiments, we used the NOAA Air Resources Laboratory Gaussian Dispersion Model (*Draxler*, 1981) to determine the minimum CH_4 release rate that our mobile method distinguished from ambient at our various plume detection distances (minimum detection distance was 11 m, maximum was 496 m). One main assumption in the model is that the emission release occurred 1 m above ground level (AGL), however it is likely that we encountered varying emission source heights, particularly between wells and facilities. We also assumed the cloud cover to be 50% on all days, and that the cloud ceiling was an average height of 6096 m. The NOAA dispersion model computed the mixing depth using the wind speed, wind direction, and weather data input to the model. Considering a dilution of 70%, and vertical and horizontal dispersion as simulated by the model under field conditions, we found that these conditions and plume concentrations corresponded to a minimum detectable limit (MDL), or release rate, of 0.59 g/s for this study. When we were very close to emission sources (< 60 m), we would have been able to detect emission rates as low as 0.065 g/s (with dilution considered). This exceeded the resolution of Brantley et al. (2014) at a similar distance, though in precision and not accuracy because the stationary techniques of Brantley et al. (2014) are designed to maximize volumetric estimation accuracy. The more precise MDL of our study is simply the consequence of being able to confidently resolve smaller concentration deviations from background using the ratio-based methods.

3.4 Methane Emission Inventory Estimates

Using MDLs for our study, we can reasonably estimate the minimum likely emissions inventory, because it is expected that infrastructural sources with larger emission rates cumulatively contribute the majority of CH_4 emissions (Frankenberg et al., 2016; Mitchell et al., 2015; Rella et al., 2015; Subramanian et al., 2015; Zavala-Araiza et al., 2015). According to a distribution of emissions at a US oil and gas site in the Four Corners region, emissions < 0.2 g/s did not significantly contribute to the overall CH_4 flux rate (Frankenberg et al., 2016). If the US study by Frankenberg et al. (2016) reflects the emission patterns in the Montney, then our mobile method was able to capture the most significant emission sources in the area.

By applying calculated emission rates to our emission incidence values for each well and facility type, we estimated the total volume of CH_4 being released annually from sites emitting at rates above our MDL. We used our MDL of 0.59 g/s to represent average emission rates from well pads in the Montney. This value is likely a conservative estimate because it is the smallest value detected at our mean detection distance (319 m), and the majority of our emission detections occurred around this value (Fig. 2). However, it should be noted that this value overestimates emissions for the (small number of) well pads with detection distances < 60 m and emission rates < 0.59 g/s. However, (Brantley et al., 2014) showed that the largest sample population of well pads measured by OTM33A (n=107) had a mean emission rate exceeding 0.59 g/s. As a result, it is reasonable to assume that our MDL serves as a reasonable average emission rate for well pads in a natural gas development, and one that allows us to estimate emission inventories for Montney well pads. For facilities, however, plumes are often emitted from higher above the ground surface, and the high concentration core of those plumes may not descend fully within a few hundred metres horizontal distance, to our 1 m AGL intake. As a result, the emissions we detected from facilities may significantly underestimate total emissions from those sources. For this reason, instead of actual measured MDLs we instead used previously-published natural gas facility emission volumes of 2.2 g/s (Omara et al., 2016), plus our frequency estimates, in order to estimate a total Montney based source inventory.

The minimum reasonable inventory is given in Table 2. Based on the types of infrastructure we surveyed, and their corresponding 50% persistence emission frequencies, we estimate that total CH_4 emissions from the wells we surveyed are 8216 tonnes per year, and total CH_4 emissions from the facilities we surveyed are 5936 tonnes per year. We therefore estimate that, in total, there are just over 14,150 tonnes per year of CH_4 emissions from all wells and facilities surveyed in this study. If we extrapolate these values to cover all natural gas wells and facilities in the BC portion of the Montney formation (using infrastructure numbers derived from BCOGC GIS database), that translates to 72,900 tonnes CH_4 per year from wells, and about 39,000 tonnes CH_4 per year from facilities, totalling more than 111,800 tonnes CH_4 per year overall (3,564,000 tonnes per year CO_2e using a 100-year GWP of 30). These measurements and estimates represent emissions from infrastructure emitting > 0.59 g/s from our average detection distance, and are therefore are representative of the more significant, higher emitting sites in the area, and not small emissions that would be detectable only at close distance on the well pad. Furthermore, our estimates did not include some well types (including cased and drilled) for which our sample size was not large enough to reliably determine emission frequency, nor did it include transport-related emissions, or emissions from well completions. For these reasons, in addition to the measurement limitation imposed by our MDL, our calculations underestimate the actual CH_4

emissions from wells. A comprehensive understanding of emissions in the BC Montney would also involve quantifying emissions below our MDL (< 0.59 g/s), potentially using on-well pad screening surveys with our vehicle, and also onsite techniques to measure smaller emissions.

From all provincial energy sector practices, BC estimates fugitive CH_4 emissions to be 78,000 tonnes per year, and stationary combustion CH_4 emissions to be 17,000 tonnes per year (British Columbia Ministry of Environment (2012)). Our estimated volume of 111,889 tonnes CH_4 per year (solely for infrastructure emitting >0.59 g/s) suggests that Montney-related natural gas activity contributes more than 117% of this total value for BC. Our calculations are therefore higher than BC's emissions estimate when we consider that natural gas production from the Montney formation was 55% of BC's total production in 2014 (BC Oil and Gas Commission, 2014) (which would be equivalent to about 52,250 tonnes per year).

Although our CH_4 emission estimates for the Montney exceed the estimates by the BC OGC, they remain lower than recent top-down oil and gas emission studies in the US. For example, in May 2014, Peischl et al. (2016) conducted airborne monitoring surveys of wells that produce more than 97% of North Dakota Bakken formation oil and gas and found that just under 250,000 tonnes of CH_4 were being emitted annually. According to North Dakota state government records, there were 10,892 producing oil and gas wells in North Dakota at the time of the surveys by Peischl et al. (2016). This means that annual CH_4 emissions were an estimated $\sim$23.0 tonnes per well. Similarly, in 2013 Karion et al. (2015) performed airborne surveys over the Barnett shale in Texas and estimated that just over 525,000 tonnes of CH_4 are released annually from this development. Texas state records show that as of early 2013 there were 16,821 producing oil and gas wells accessing the Barnett shale formation, which means that annual CH_4 emissions in this development were $\sim$31.3 tonnes per well. The analogous figure in the Montney is $\sim$7.3 tonnes per well. The lower emissions per well in the BC Montney are consistent with the relatively low incidence of excess atmospheric CH_4 in the region on all surveys compared to higher atmospheric CH_4 values recorded in US developments.

4 Conclusion

Unconventional natural gas development in the BC Montney began less than a decade ago, and so the majority of infrastructure is new in comparison to many old conventional oil developments in Alberta and Saskatchewan. Though the Montney is regarded as a young development, there are many locations where old, decommissioned infrastructure exists, and in a generally unkept state. Our results show that older infrastructure is more prone to persistent leaks, albeit at similarly low eCH_4 severity in comparison to younger wells. These results reinforce the need for regulators to pay attention not only to modern equipment, but also legacy wells and infrastructure.

In calculating the frequency of emissions in the BC Montney above our MDL of 0.59 g/s, we found that about 47% of active wells were emitting. Furthermore, abandoned and aging infrastructure were regularly associated with emissions. The emissions we detected from facilities were consistent in both presence and eCH_4 severity, however our mobile detection method is sensitive to plume transport turbulence associated with emissions higher above ground level such as flare stacks.

Our calculated emission frequency values, combined with estimated and pre-established emission factors for wells and facilities, provided a CH_4 emission volume estimate of more than 111,800 tonnes per year for the BC portion of the Montney.

This value exceeds the province-wide estimate provided by the government of British Columbia even though the Montney only represents about 55% of BC's total natural gas production. But, in comparison to studies at select US natural gas sites (Peischl et al., 2016; Karion et al., 2015), our results suggest that natural gas activity in the Montney formation may emit both less frequently and less severely than US comparators.

Methane emission reduction strategies for large natural gas developments such as the Montney should focus on first locating super-emitting sites, and then following up with site-specific emission techniques such as FLIR cameras. This strategy would support LDAR already in place, in a way that would minimize cost to individual operators. Our results show that a mobile surveying approach for large developments such as the Montney can help to locate probable emitting infrastructure pieces that contribute to the heavy-tailed emission distribution found by Frankenberg et al. (2016). Also, using a mobile survey method to define persistent emitting infrastructure allows for the probable emission type (consistent or episodic) to be deduced. Our study highlights the need for emission reduction efforts in the Montney to be focused on the higher-emitting to super-emitting production wells, as well as abandoned, and aging infrastructure in this natural gas development.

5 Data availability

Datasets of atmospheric gas concentrations, wind, and temperature data are available upon request. Oil and gas infrastructure location data can be accessed through the BC Oil and Gas Commission Open Data Portal (BC Oil and Gas Commission, n.d.)

Author contributions. D. Risk and M. Lavoie developed the algorithms for background subtraction and plume detection. E. Atherton designed the field campaigns with insight from J. Werring. E. Atherton, J. Werring, A. Marshall, J.P. Williams, and C. Minions carried out the field surveys. The data were compiled and analyzed by E. Atherton with help from M. Lavoie and C. Fougere. The manuscript was prepared by E. Atherton and D. Risk.

References

Allen, D.T., Torres, V.M., Thomas, J., Sullivan, D.W., Harrison, M., Hendler, A., Herndon, S.C., Kolb, C.E., Fraser, M.P., Hill, A.D., Lamb, B.K., Miskimins, J., Sawyer, R.F., Seinfeld, J.H.: Measurements of methane emissions at natural gas production sites in the United States. P. Natl. Acad. Sci. USA. 110, 17768-17773. doi:10.1073/pnas.1304880110, 2013.

Alvarez, R.A., Pacala, S.W., Winebrake, J.J., Chameides, W.L., Hamburg, S.P.: Greater focus needed on methane leakage from natural gas infrastructure. P. Natl. Acad. Sci. USA. 109, 6435-6440. doi:10.1073/pnas.1202407109, 2012.

BC Oil and Gas Commission: Montney Formation Play Atlas NEBC. Available at: https://www.bcogc.ca/montney-formation-play-atlas-nebc, 2012.

BC Oil and Gas Commission: Energy Briefing Note: The Ultimate Potential for Unconventional Petroleum from the Montney Formation of British Columbia and Alberta. Available at: https://www.neb-one.gc.ca/nrg/sttstc/ntrlgs/rprt/ltmtptntlmntnyfrmtn2013/ltmtptntlmntnyfrmtn2013-eng.pdf, 2013.

BC Oil and Gas Commission: Hydrocarbon and By-Product Reserves in British Columbia. Available at: https://www.bcogc.ca/node/12952/download, 2014.

BC Oil and Gas Commission Open Data Portal. Last access: 20 July, 2015. Available at: http://data.bcogc.opendata.arcgis.com/, n.d..

British Columbia Ministry of Environment: British Columbia Greenhouse Gas Inventory Report. Available at: http://www2.gov.bc.ca/assets/gov/environment/climate-change/reports-and-data/provincial-ghg-inventory-report-bcs-pir/pir-2012-full-report.pdf, 2012.

Beaubien, S.E., Jones, D.G., Gal, F., Barkwith, A.K. a. P., Braibant, G., Baubron, J.-C., Ciotoli, G., Graziani, S.asdf Lister, T.R., Lombardi, S., Michel, K., Quattrocchi, F., Strutt, M.H.: Monitoring of near-surface gas geochemistry at the Weyburn, Canada, CO2-EOR site, 2001-2011. Int. J. Greenh. Gas Con. 16, S236-S262. doi:10.1016/j.ijggc.2013.01.013, 2013.

Brantley, H.L., Thoma, E.D., Squier, W.C., Guven, B.B., Lyon, D.: Assessment of Methane Emissions from Oil and Gas Production Pads using Mobile Measurements. Envir. Sci. Tech. 48, 14508-14515. doi:10.1021/es503070q, 2014.

Burnham, A., Han, J., Clark, C.E., Wang, M., Dunn, J.B., Palou-Rivera, I.: Life-Cycle Greenhouse Gas Emissions of Shale Gas, Natural Gas, Coal, and Petroleum. Envir. Sci. Tech. 46, 7430-7430. doi:10.1021/es301020p, 2012.

Canadian Natural Gas Initiative: Life Cycle Greenhouse Gas Emissions: a literature review of key studies comparing emissions from natural gas and coal. Available at: http://www.capp.ca/responsible-development/air-and-climate/greenhouse-gas-emissions, 2012.

Caulton, D.R., Shepson, P.B., Santoro, R.L., Sparks, J.P., Howarth, R.W., Ingraffea, A.R., Cambaliza, M.O.L., Sweeney, C., Karion, A., Davis, K.J., Stirm, B.H., Montzka, S.A., Miller, B.R.: Toward a better understanding and quantification of methane emissions from shale gas development. P. Natl. Acad. Sci. USA. 111, 6237-6242. doi:10.1073/pnas.1316546111, 2014.

Draxler, R.: Forty-Eight Hour Atmospheric Dispersion Forecasts at Selected Locations in the United States NOAA Technical Memorandum ERL ARL-100. Web model available at: https://www.ready.noaa.gov/READY_gaussian.php, 1981.

Frankenberg, C., Thorpe, A.K., Thompson, D.R., Hulley, G., Kort, E. A., Vance, N., Borchardt, J., Krings, T., Gerilowski, K., Sweeney, C., Conley, S., Bue, B.D., Aubrey, A. D., Hook, S., Green, R. O.: Airborne methane remote measurements reveal heavy-tail flux distribution in Four Corners region. P. Natl. Acad. Sci. USA. 113, 9734-9739. doi:10.1073/pnas.1605617113, 2016.

Govindan, R., Korre, A., Durucan, S., Imrie, C.E.: A geostatistical and probabilistic spectral image processing methodology for monitoring potential CO2 leakages on the surface. Int. J. Greenh. Gas Con. 5, 589-597. doi:10.1016/j.ijggc.2010.04.014, 2011.

Howarth, R.W., Santoro, R., Ingraffea, A.: Methane and the greenhouse-gas footprint of natural gas from shale formations. Climatic Change. 106, 679-690. doi:10.1007/s10584-011-0061-5, 2011.

Hultman, N., Rebois, D., Scholten, M., Ramig, C.: The greenhouse impact of unconventional gas for electricity generation. Environ. Res. Lett. 6, 044008. doi:10.1088/1748-9326/6/4/044008, 2011.

Hurry, J., Risk, D., Lavoie, M., Brooks, B.G., Phillips, C.L., Gockede, M.: Atmospheric monitoring and detection of fugitive emissions for Enhanced Oil Recovery. Int. J. Greenh. Gas Con. 45, 1-8. doi:10.1016/j.ijggc.2015.11.031, 2016.

Intergovernmental Panel on Climate Change: Intergovernmental Panel on Climate Change: Fifth Assessment Report; Geneva. 2014.

Jiang, M., Griffin, M., Hendrickson, C., Jaramillo, P., VanBriesen, J., and Venkatesh, A.: Life cycle greenhouse gas emissions of Marcellus shale gas. Envir. Res. Lett. 6. doi:10.1088/1748-9326/6/3/034014, 2011.

Karion, A., Sweeney, C., Petron, G., Frost, G., Hardesty, R.M., Kofler, J., Miller, B.R., Newberger, T., Wolter, S., Banta, R., Brewer, A., Dlugokencky, E., Lang, P., Montzka, S.A., Schnell, R., Tans, P., Trainer, M., Zamora, R., Conley, S.: Methane emissions estimate from airborne measurements over a western United States natural gas field. Geophys. Res. Lett. 40, 4393-4397. doi:10.1002/grl.50811, 2013.

Karion, A., Sweeney, C., Kort, E.A., Shepson, P.B, Brewer, A., Cambaliza, M., Conley, S.A., Davis, K., Deng, A., Hardesty, M., Herndon, S.C., Lauvaux, T., Lavoie, T., Lyon, D., Newberger, T., Petron, G., Rella, C., Smith, M., Wolter, S., Yacovitch, T.I., Tans, P.: Aircraft-Based Estimate of Total Methane Emissions from the Barnett Shale Region. Envir. Sci. Tech. 49, 8124-8131. doi:10.1021/acs.est5b00217, 2015.

Lyon, D.R., Zavala-Araiza, D., Alvarez, R.A., Harriss, R., Palacios, V., Lan, X., Talbot, R., Lavoie, T., Shepson, P., Yacovitch, T.I., Herndon, S.C., Marchese, A.J., Zimmerle, D., Robinson, A.L., Hamburg, S.P.: Constructing a Spatially Resolved Methane Emission Inventory for the Barnett Shale Region. Envir. Sci. Tech. 49, 8147-8157. doi:10.1021/es506359c, 2015.

Mayer, B., Shevalier, M., Nightingale, M., Kwon, J.-S., Johnson, G.,E Perkins, E.: Tracing the movement and the fate of injected CO_2 at the IEA GHG Weyburn-Midal CO_2 Monitoring and Storage project (Saskatchewan, Canada) using carbon isotope ratios. Int. J. Greenh. Gas Con., 16S, S177-S184. doi: 10.1016/j.ijggc.2013.01.035, 2013.

Mitchell, A.L., Tkacik, D.S., Roscioli, J.R., Herndon, S.C., Yacovitch, T.I., Martinez, D.M., Vaughn, T.L., Williams, L.L., Sullivan, M.R., Floerchinger, C., Omara, M., Subramanian, R., Zimmerle, D., Marchese, A.J., Robinson, A.L.: Measurements of Methane Emissions from Natural Gas Gathering Facilities and Processing Plants: Measurement Results. Envir. Sci. Tech. 49, 3219-3227. doi:10.1021/es5052809, 2015.

National Energy Board: Canada's Energy Future 2016: Energy Supply and Demand Projections to 2040. Available at: https://www.neb-one.gc.ca/nrg/ntgrtd/ftr/2016/ 2016nrgftr-eng.pdf, 2016.

National Energy Technology Laboratory: Cost and performance baseline for fossil energy plants, Volume 1: Bituminous coal and natural gas to electricity. Revision 2. November. DOE/NETL-2010/1397. United States Department of Energy. 2010.

Omara, M., Sullivan, M.R., Li, X., Subramanian, R., Robinson, A.L., Presto, A.A.: Methane Emissions from Conventional and Unconventional Natural Gas Production Sites in the Marcellus Shale Basin. Envir. Sci. Tech. 50, 2099-2107. doi:10.1021/acs.est.5b05503, 2016.

Peischl, J., Karion, A., Sweeney, C., Kort, E.A., Smith, M.L., Brandt, A.R., Yeskoo, T., Aikin, K.C., Conley, S.A., Gvakharia, A., Trainer, M., Wolter, S., Ryerson, T.B.: Quantifying atmospheric methane emissions from oil and natural gas production in the Bakken shale region of North Dakota. J. Geophys. Res-Atmos. 121, 2015JD024631. doi:10.1002/2015JD02463, 2016.

R Core Team (2016). R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. URL http://www.R-project.org/.

Rella, C.W., Tsai, T.R., Botkin, C.G., Crosson, E.R., Steele, D.: Measuring Emissions from Oil and Natural Gas Well Pads Using the Mobile Flux Plane Technique. Envir. Sci. Tech. 49, 4742-4748. doi:10.1021/acs.est.5b00099, 2015.

Rich, A., Grover, J.P., Sattler, M.L.: An exploratory study of air emissions associated with shale gas development and production in the Barnett Shale. JAPCA J. Air Waste Ma. 64, 61-72. doi:10.1080/10962247.2013.832713, 2014.

Romanak, K.D., Bennett, P.C., Yang, C., Hovorka, S.D.: Process-based approach to CO2 leakage detection by vadose zone gas monitoring at geologic CO2 storage sites. Geophys. Res. Lett. 39, L15405. doi:10.1029/2012GL052426, 2012.

Schneising, O., Burrows, J.P., Dickerson, R.R., Buchwitz, M., Reuter, M., Bovensmann, H.: Remote sensing of fugitive methane emissions from oil and gas production in North American tight geologic formations. Earth Future. 2, 548-558. doi:10.1002/2014EF000265, 2014.

Skone, T.J.: Life Cycle Greenhouse Gas Inventory of Natural Gas Extraction, Delivery and Electricity Production. National Energy Technology Laboratory, U.S. Department of Energy, DOE/NETL-2011/1522, 2011.

Stephenson, T., Valle, J.E., Riera-Palou, X.: Modeling the Relative GHG Emissions of Conventional and Shale Gas Production. Envir. Sci. Tech. 45, 10757-10764. doi:10.1021/es2024115, 2011.

Subramanian, R., Williams, L. L., Vaughn, T. L., Zimmerle, D., Roscioli, J. R., Herndon, S. C., Yacovitch, T. I., Floerchinger, C., Tkacik, D. S., Mitchell, A. L.: Methane Emissions from Natural Gas Compressor Stations in the Transmission and Storage Sector: Measurements and Comparisons with the EPA Greenhouse Gas Reporting Program Protocol. Envir. Sci. Tech. 49 (5), 3252-3261. doi:10.1021/es5060258, 2015.

Zavala-Araiza, D., Lyon, D.R., Alvarez, R.A., Davis, K.J., Harriss, R., Herndon, S.C., Karion, A., Kort, E.A., Lamb, B.K., Lan, X., Marchese, A.J., Pacala, S.W., Robinson, A.L., Shepson, P.B., Sweeney, C., Talbot, R., Townsend-Small, A., Yacovitch, T.I., Zimmerle, D.J., Hamburg, S.P.: Reconciling divergent estimates of oil and gas methane emissions. P. Natl. Acad. Sci. USA. 112, 15597-15602. doi:10.1073/pnas.1522126112, 2015.

Table 1. Survey statistics by Route. Route locations are shown in Figure 1.

Routes	Control	Regional	1	2	3	4	All
Route Length (km)	370	545	145	210	235	280	1785
Number of Repeat Surveys	3	3	6	6	6	6	30
Total km Surveyed	1110	1635	870	1260	1410	1680	7965
Unique Sampled Wells	152	436	172	241	298	182	1481
Unique Sampled Facilities	10	113	63	29	34	16	265
Unique Sampled Groups	49	304	146	88	110	51	748

Table 2. Emission volume calculations for all surveyed infrastructure, and also extrapolated to account for all wells and facilities within the BC portion of the Montney formation. Our minimum detection limit (MDL) of 0.59 g/s was used as the emission factor for wells. Facility emission volumes are from Omara et al. (2016) because our sampling from facilities was probabilistic due to emission height variance.

Type	Emission n	Emission Freq (%)	Emission Volume (tonnes/year)	Emission Total (tonnes/year)
Surveyed Wells				
Active	676	47	18.6	5910
Abandoned	228	26	18.6	1103
Cancelled	130	35	18.6	846
Completed	64	30	18.6	357
Surveyed Facilities	265	32	70	5936
Total CH_4 volume				14152
Montney Wells				
Active	5294	47	3.2	46,280
Abandoned	2149	26	3.2	10,392
Cancelled	1989	35	3.2	12,948
Completed	582	30	3.2	3248
Montney Facilities	1742	32	70	39021
Total CH_4 volume				111,889

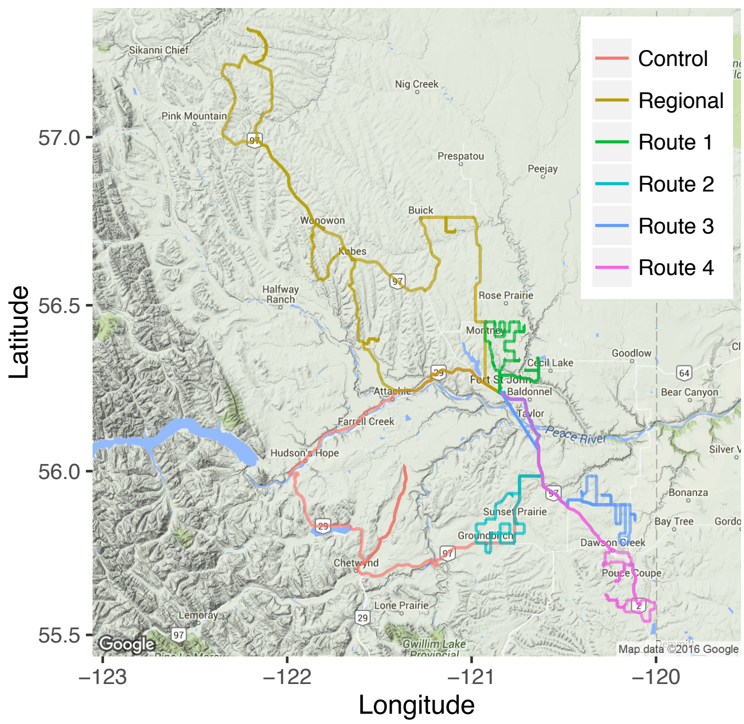

Figure 1. Map of mobile surveying routes. Each route was surveyed six times in August - September, 2015. The Regional Route and Routes 2-4 dissected unconventional natural gas developments. Route 1 surveyed conventional oil. The Control Route was located in an area with a comparatively small amount of oil and gas development, although due to lack of accessible roads in the area it passed by some infrastructure on Route 2 upon returning to the Fort St. John area.

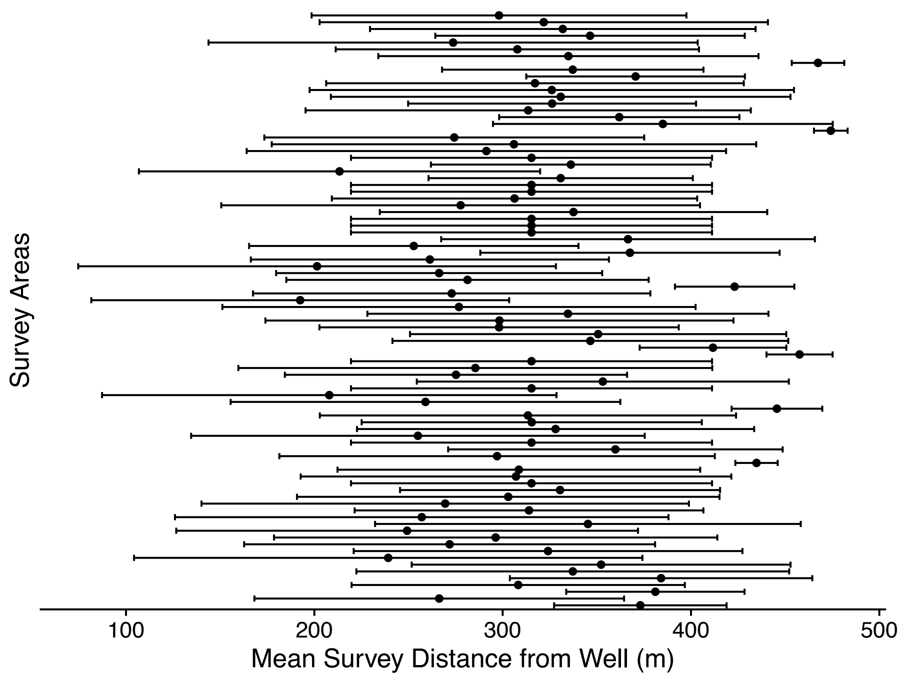

Figure 2. Mean distance from infrastructure while surveying in each of 88 industry-defined areas of the Montney that we accessed. One standard deviation from the mean shows the range of distances at which we were sampling downwind of infrastructure.

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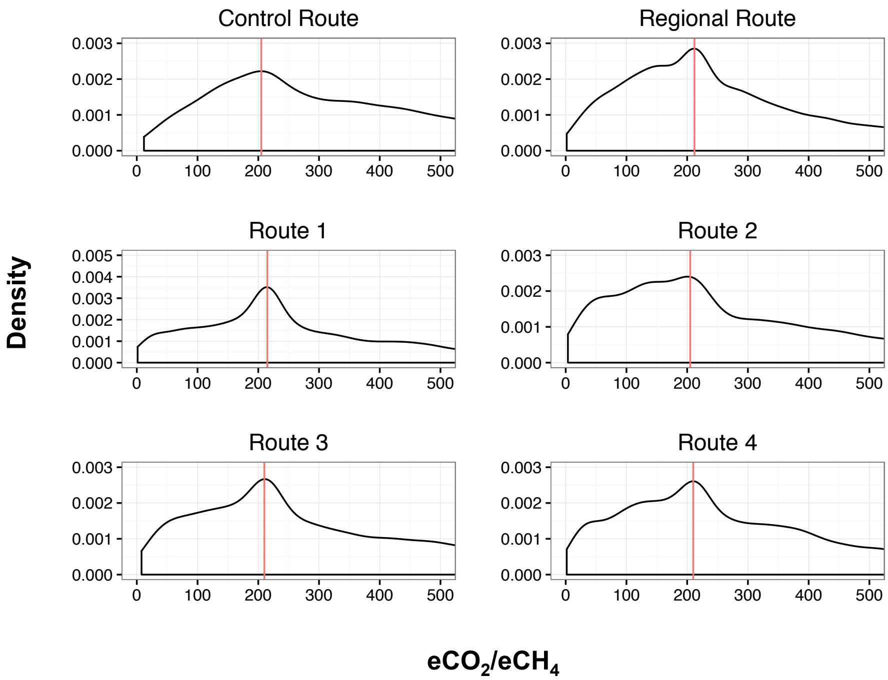

Figure 3. Kernel density plots showing the density of eCO_2:eCH_4 measurements on each route. Red vertical lines indicate natural eCO_2:eCH_4 values about 215. Methane-enriched peaks are visible to the left of the natural ratio on all routes except for the Control, where the slope approaches zero with no peaks because substantially less natural gas infrastructure was surveyed. Ratios higher than the natural represent CO_2-rich plumes which would not be caused by natural gas related emissions, but likely diluted car exhaust fumes, or other industry types.

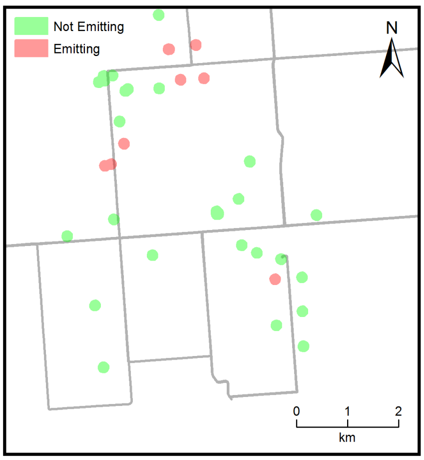

Figure 4. A subset of spatial data in its raw attributed form, extracted from 1 of 30 surveys. In this case 31 wells or facilities were surveyed, though throughout the whole study almost 1500 wells were surveyed (in triplicate). Using spatial data, along with well or facility characteristics, we were able to infer drivers of emissions in our study area.

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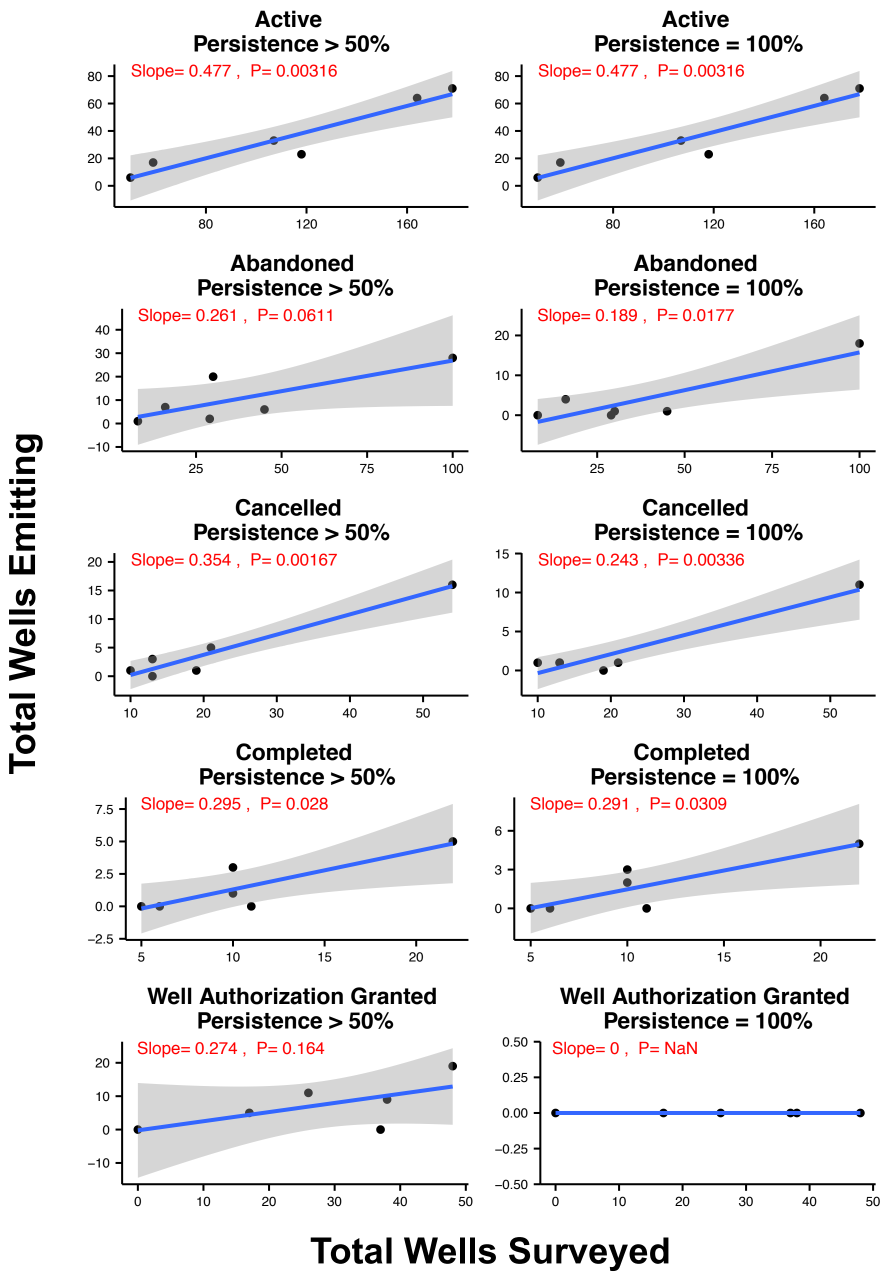

Figure 5. Well pad emission frequencies for active, suspended, abandoned, and other categories. Each datapoint represents the frequency found on all passes of a single route. Wells were tagged as emitting only when they met the geochemical, geospatial, and persistence criteria. Slopes represent the increase in emissions incidence with the increase in number of wells surveyed.

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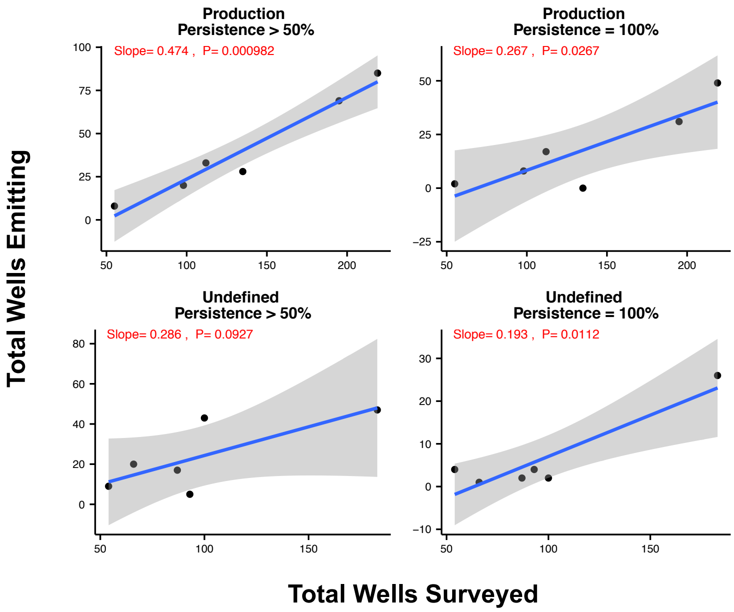

Figure 6. Well pad emission frequency for active wells only. As seen by the slope, most of the well emissions are driven by the class of active wells. Almost half of active in-production well pads met our criteria for probable emitters.

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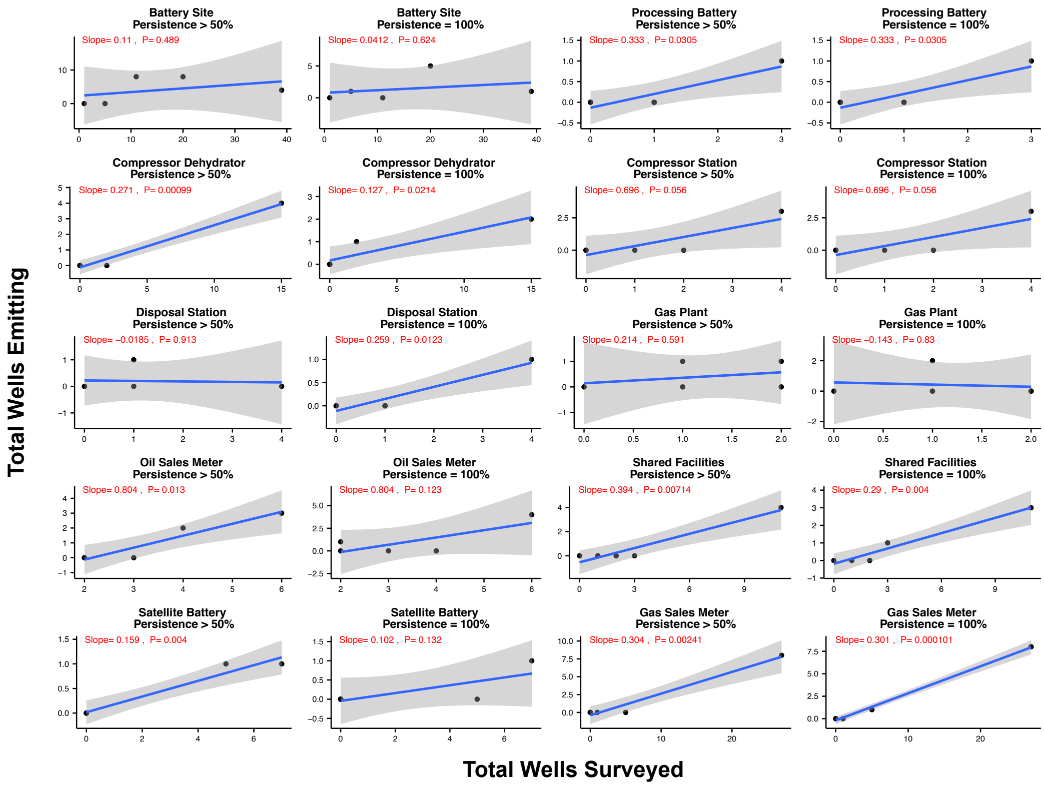

Figure 7. Emission frequencies for all classes of facilities surveyed. Each datapoint represents the frequency found on all passes of a single route. Facilities were tagged as emitting only when they met the geochemical, geospatial, and persistence criteria. Slopes represent the increase in emissions incidence with the increase in number of facilities surveyed.

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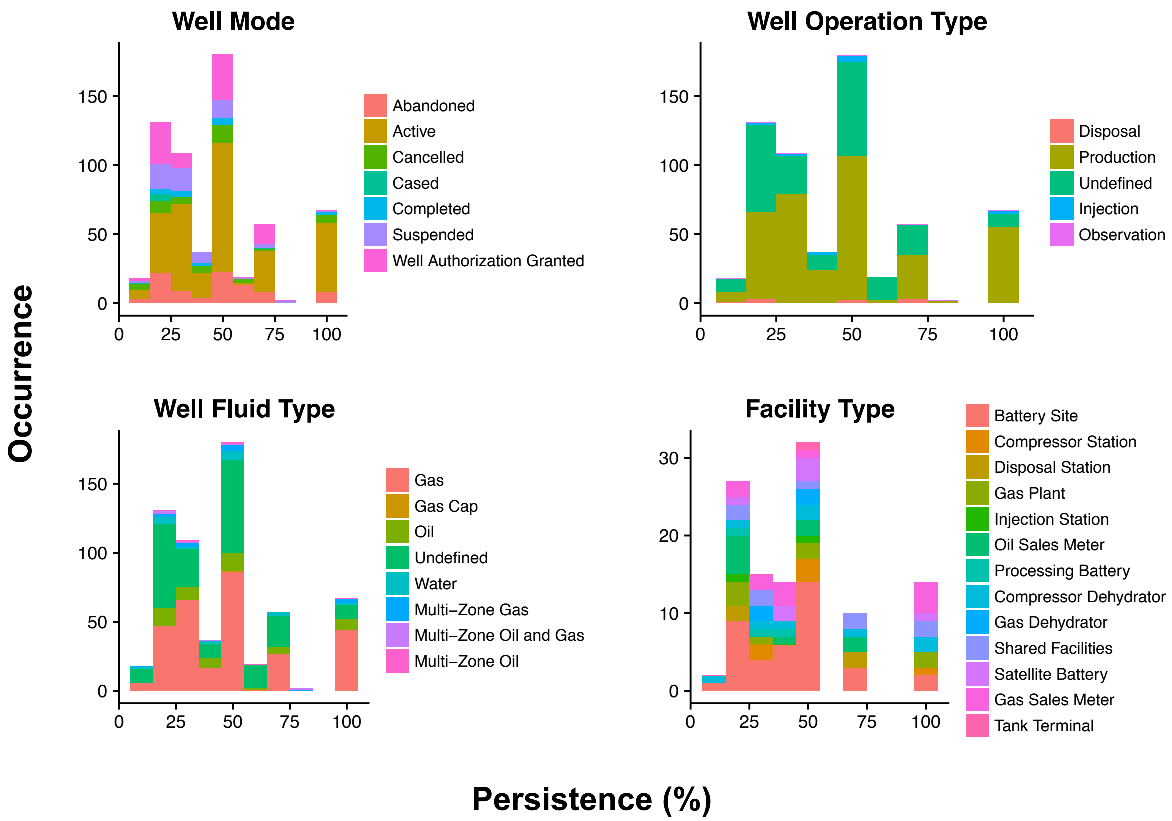

Figure 8. Occurrence (number) vs emission Persistence (%) across surveys. Persistence refers to the repeated tagging of the infrastructure according to criteria on each of the passes when our truck was downwind and could have potentially detected an emission from the infrastructure in question.

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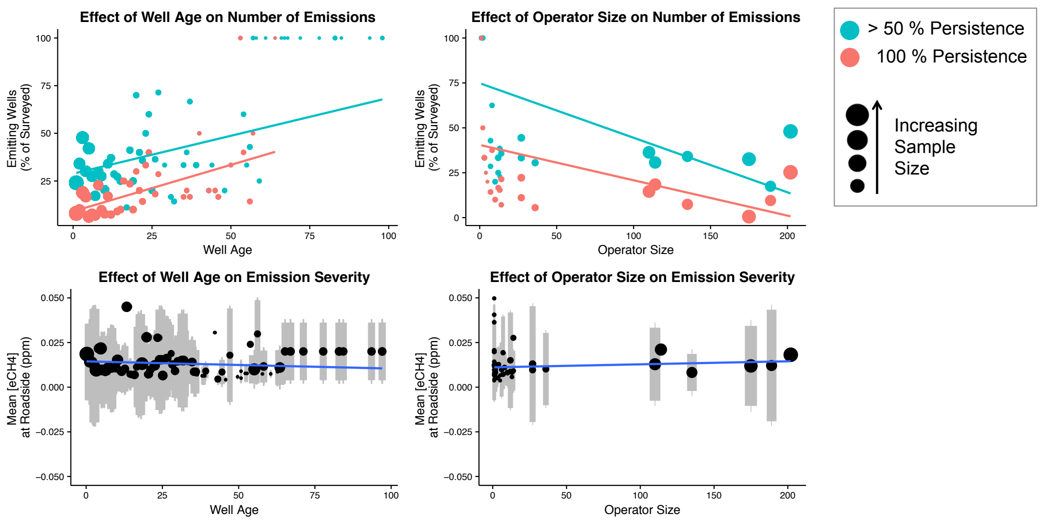

Figure 9. Effect of age and operator size on detected emissions. The size of the dots represents number of samples. Red dots are those recorded at the 100% persistence level, green dots are at 50% persistence. The grey error bars in the bottom two plots are one standard deviation of the mean of 1) all anomalous datapoints in the radius of wells of each age bin (binned by each year); or, 2) all anomalous datapoints in the radius of each operators' wells.

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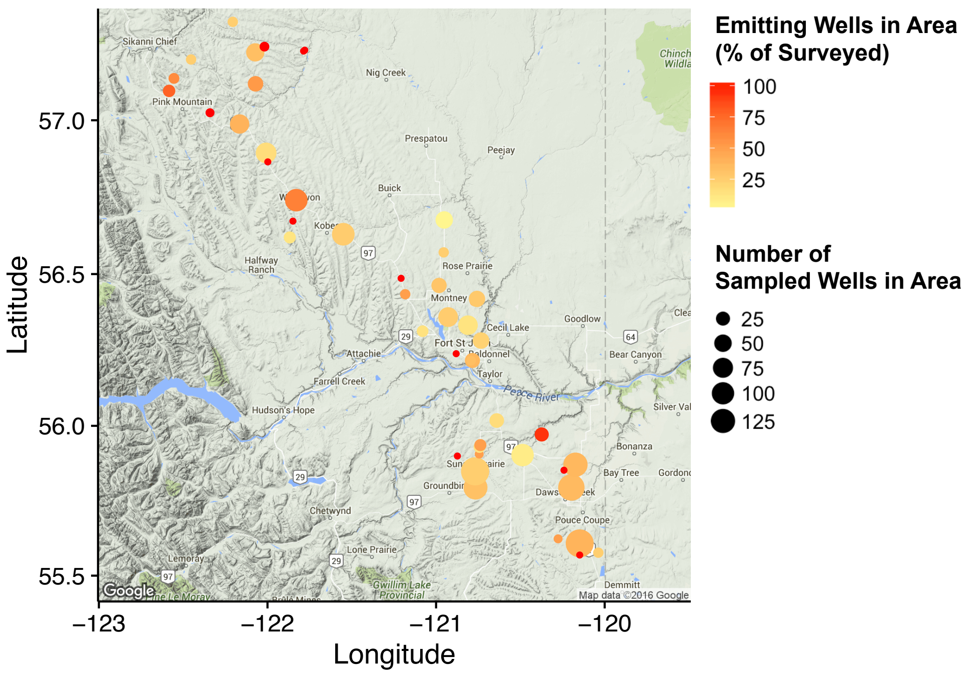

Figure 10. Distribution of emitting infrastructure by industry-defined area. The size of the circles represents the number of measurements we took downwind from individual wells or facilities in each area. The colour of the circles represents the frequency of emitting infrastructure in each area.