Research Project **ES-Wells-2023-01** Technical Report

Evaluating Fugitive Methane Oxidation in Natural Soils of British Columbia through Data Analyses, Field Investigation and Reactive Transport Modelling

Dr. Aaron Cahill

Institute of Geoenergy Engineering and the Lyell Centre, Heriot Watt University, Edinburgh, UK

*corresponding author: a.cahill@hw.ac.uk

With input from Jordan Van Besouw (BC ER) and Leah Davies (IOL)

Executive Summary

British Columbia has an extensive onshore oil and gas industry that hosts ~24,000 energy wells. A small but growing number of BC wells are identified as suffering well integrity failure and releasing fugitive gas (formed primarily by methane; CH_4) into the environment through soils around the well. During reactive transport through soils, a portion of fugitive CH_4 can be aerobically oxidized via microbes to form CO_2 resulting in a mixture of CH_4 and CO_2 reaching ground surface and emitting to atmosphere. The process of microbially-mediated aerobic CH_4 oxidation in soils has become of increasing interest in recent years as it reduces the total amount of CH_4 being directly released into the atmosphere, offering a 98% reduction in GHG emissions (i.e., in terms of CO_2 equivalents) for every mole of CH_4 converted to CO_2 (considering a 20-year period). Here, we examined CH_4 oxidation in natural soils in BC, through a comprehensive scientific investigation with a view to informing regulatory policy and developing potential solutions that can mitigate GHG emissions from integrity-compromised wells. The research involved three synergistic work packages (WP1 – 3) advanced sequentially with their activity and outcomes described as follows.

In WP1, existing BC related field data relevant to CH_4 oxidation in soils at and around oil and gas wells was collated and analyzed with a view to appraising occurrence, extents and rates. More specifically, data from three key studies was attained and re-evaluated showing that CH_4 oxidation in soils appears ubiquitous in BC and often occurs to a great extent; typically converting >50% of leaking CH_4 to CO_2. Estimated oxidation rates from these data ranged from 2 g CH_4/m^2 of soils/day, to as high as 144 g CH_4/m^2 of soils/day, with an average of ~30 g CH_4/m^2/day. It was noted that the rate of CH_4 oxidation in soils around leaking oil and gas wells is strongly controlled by leakage rate. Here, the data suggests that increasing leakage rates lead to increasing oxidation rates and that leakage rates of up to 10 m^3 (@STP) or 7 kg per day of CH_4 might comfortably be broken down by soils around leaking oil and gas wells with minimal engineering.

In WP2, new and uniquely designed field investigations focusing on wells sites where integrity failure was suspected to be occurring and CH_4 was assumed as being released into natural soils were advanced. In particular, wellsite WA22800, at which significant levels of soils based CH_4 oxidation was previously identified, formed a key focus. New observations were consistent with previously conducted fieldwork showing clearly the presence of anomalous CH_4 and CO_2 emissions at wellsite WA22800 and anomalously elevated CO_2 emissions at the other wellsites examined (i.e. with no excess or anomalous CH_4 present). Aside from general consistency in observed emissions profiles between years, wellsite WA22800 exhibited 10-fold higher CH_4 fluxes this year compared to last, while an relatively high average oxidation rate for CH_4 of ~230 g of CH_4 /m^2 of soils/day was estimated

from newly collected field data. Meanwhile a profound difference in soil microbiome between soils at the leaking well head and background locations was observed, whereby a consortia of bacteria including microorganisms in the families of Methylomonadaceae, Beijerinckiaceae and Desulfocapsaceae were identified as likely to be involved in the CH_4 oxidation process.

In WP3, reactive transport of fugitive CH_4 through soils, including surficial emissions, were modelled using the USDA CALMIM model. Here, modelling highlighted that extents and rates of CH_4 oxidation in soils and therefore emissions profiles are driven by climatic conditions (i.e. primarily by prevailing temperature with shorter-term influence by precipitation), soil properties (i.e. type, thickness, maximum potential oxidation rates) and methane leakage rate (i.e. incorporated in the model as CH_4 concentration at depth). We demonstrated that large swings in air temperature across the year (i.e. from an average of -15 °C in winter to 15 °C in summer) lead to CH_4 oxidation and emissions predictions that vary by at least a factor of ten over an annual cycle. In our simulations, very high rates of oxidation coupled to very low or no methane emissions were modelled during the summer months, while a cessation in methane oxidation coupled with emissions > 100 g/m^2/day were modelled during winter months. Overall, these simulations suggest that it is vital to incorporate climatic conditions into predictions of CH_4 oxidation extents and rates in order to accurately predict annual emissions and appraise overall mass oxidized. In our case, if we extrapolate the field measured August oxidation rate to the whole year, a total of 12.6 kg of CH_4/m^2 is estimated to be released into the atmosphere. However, by including climatic variability and soil transport properties in CALMIM, we reach an annual estimate three times larger (i.e. 36.15 kg of CH_4/m^2). This is due to CH_4 being converted to CO_2 at a much lower rates in lower temperature months, whereby at the coldest times no CH_4 oxidation occurs at all and all fugitive gas is directly emitted to atmosphere.

Overall, we find that oxidation of CH_4 in natural soil systems is likely ubiquitous, occurs inherently and is already mitigating emissions at integrity compromised oil and gas wells in BC and elsewhere. Moreover, we conclude that various factors control rates and extents of CH_4 oxidation and more work is needed to understand their interplay and identify optimal conditions for the process to occur. This finding infers overall extents and benefits of CH_4 oxidation in soils around a leaking well will vary well to well and seasonally at given well. Based on this, we suggest that some minimal site preparation pre- or post-leakage could enhance and optimize soils based CH_4 oxidation in BC, and highlight that natural soils based CH_4 oxidation could also be used to mitigate SCVF during a wells active life. Finally, we identified a consortia of micro-organisms driving soil based CH_4 oxidation and more understanding on their functioning and interplay is needed to further optimize and potentially enhance the process if it is to be used as an integrity compromised well management approach.

Contents

Introduction and Context

British Columbia has an extensive onshore oil and gas industry that has seen some 24,000 energy wells constructed in the province over the past 100 years[1,2]. Unfortunately, a small but growing number of these wells are being identified as suffering well integrity failure during their active and post-abandonment lives[2-4]; a challenge that will need to be managed in the coming years[5]. Well integrity failure involves the break down of an energy well barrier system, leading to either inside casing leakage (ICL) and surface casing vent flow (SCVF) and/or outside casing leakage (OCL) and gas migration (GM). In the case of ICL and SCVF, as the vent is typically open, fugitive natural gas comprised primarily of methane (CH_4) is emitted directly to atmosphere, contributing to global heating with 86 times more impact than emitting CO_2 alone over a 20-year period[6]. In the case of OCL and GM, fugitive gas is typically released at some depth below ground into soils and sediments intersected by the well and reactively transported towards the ground surface due to buoyancy forces[2]. During reactive transport through the soils, a portion of fugitive CH_4 can be aerobically oxidized via microbes to form CO_2 resulting in a mixture of CH_4 and CO_2 reaching ground surface and emitting to atmosphere[7,8]. Such greenhouse gas (GHG) emissions are well demonstrated by considering data from a recent study conducted in August 2022 (i.e. precursor BC OGRIS project ES-Wells-2021-02), which identified a plugged and abandoned well on agricultural land near Fort St. John, BC emitting CH_4 into the soils above the well. Field data also revealed a greater magnitude of CO_2 co-emitting to atmosphere, clear evidence for a significant degree of CH_4 oxidation occurring in the soils at the site (Figure 1). The process of microbially mediated aerobic CH_4 oxidation in soils has become of increasing interest in recent years as it reduces the total amount of CH_4 being directly released into the atmosphere, offering a 98% reduction in GHG emissions (i.e., in terms of CO_2 equivalents) for every mole of CH_4 converted to CO_2 (considering a 20-year period). The process of CH_4 oxidation in soils has long been recognized as a key biogeochemical process in the field of agricultural and soil sciences[9-11], while use of the phenomenon to reduce GHG emissions in engineered systems has also been investigated in other contexts for several decades. This is particularly true in the context of landfill "biocovers", many forms of which have been designed and deployed in various configurations (and extents of engineering) to oxidize CH_4 and reduce GHG emissions into the atmosphere [12-15].

Clearly, there is huge potential for CH_4 oxidation to form an alternative approach to full re-abandonment or expensive remedial treatments for energy wells suffering integrity failure and releasing fugitive gas. In particular, the use of natural soils at and around a well (as opposed to engineered or controlled systems) could offer a low-cost, low-tech approach that is particularly attractive in the context of integrity compromised energy wells, many of which may exist in remote locations. At such sites, standard approaches (i.e., well re-entry, cement squeezes and/or re-abandonment) have significant costs, environmental risks, and large carbon footprints. Meanwhile, engineered CH_4 oxidation systems (such as those employed at landfills) could be prohibitively expensive and complex to deploy and manage. Therefore, natural soils based CH_4 oxidation offers a particularly attractive low-tech, low cost alternative to re-abandonment/remedial treatment, at least where fugitive gas release rates are low or marginal (although how low is unclear).

Despite recent research and the clear promise that CH_4 oxidation in natural soil structures could form a low-cost, low-tech option for managing integrity failure at a large number of remotely located oil and gas wells, critical knowledge gaps persist. In fact, little research has been conducted on CH_4 oxidation in natural soil structures (within BC or elsewhere) with focus on its potential for GHG mitigation at integrity compromised oil and gas wells.

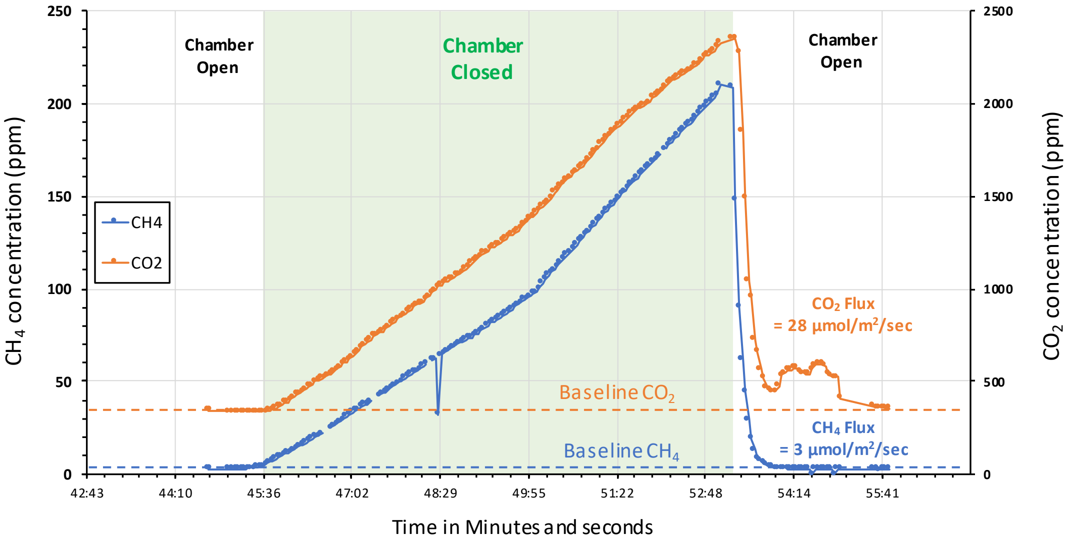


Figure 1: CH_4 and CO_2 concentrations observed during a flux measurement at a wellsite identified as suffering potential integrity failure in precursor BC OGRIS project ES-Wells-2021-02. Results show co-emission of CH_4 and CO_2 suggesting a significant level of CH_4 oxidation is occurring in these natural soils. These data prompted proposal of this follow up study to better understand the phenomena and its potential significance with respect to fugitive gas release and potential mitigation and management strategies.

While it is clear **engineered** systems could be constructed and maintained that may have significant capacity for CH_4 oxidation (e.g. such as those used at landfills), much less is understood on the ability of **natural** soils to oxidize CH_4 in non- or minimally engineered systems. Consequently, key questions associated with CH_4 oxidation in natural soils around oil and gas wells persist, including:

- How and where has methane oxidation in natural soils around integrity compromised wells been characterized or evaluated and what does existing data show?
- What factors control natural methane oxidation in soils around integrity-compromised wells?
- What rates and extents of CH_4 oxidation can occur in natural soils around integrity-compromised wells and how effective could the process be at mitigating GHG emissions to atmosphere?
- Could CH_4 oxidation in natural soils around integrity-compromised wells be used to mitigate GHG emissions/as an alternative remedial method and what might such a natural solution look like?

Here, we sought to address these key questions regarding CH_4 oxidation in natural soils, through a comprehensive scientific investigation. During this study, the potential for CH_4 oxidation in BC relevant natural soils was evaluated with a view to informing regulatory policy and developing potential alternative solutions that can mitigate GHG emissions from integrity-compromised wells.

Project Structure, Aims and Objectives

The project involved three synergistic work packages (WP1 – 3) advanced sequentially over 12 months (commencing May 1st 2023), including:

- **WP1: Evaluation of CH_4 oxidation in natural soils using existing data and literature review**
- **WP2: Field investigations examining CH_4 oxidation in natural soils of BC**
- **WP3: Reactive Transport Modelling and Remedial Potential Appraisal and Outline**

Overall, this project sought to systematically and rigorously evaluate CH_4 oxidation in natural soils at and around integrity compromised oil and gas wells in BC using; a) existing data, b) collecting new data, and, c) conducting data informed reactive transport modeling.

This technical report is divided into sections that outline the specific activities, results and findings from each of the individual work packages (i.e., WP1 – 3). After describing each work package individually, the findings are used to explore if and how natural soils based CH_4 oxidation might be used as a well integrity failure management strategy, before recommendations for next steps are made.

WP1: Evaluation of CH_4 Oxidation in Natural Soils Using Existing Data

1. Introduction

Many studies evaluating gas migration in soils around integrity compromised oil and gas wells have been conducted in the last decade, however, typically such studies do not evaluate collected data with respect to CH_4 oxidation in soils, or in the context of assessing potential for the process to be used to mitigate GHG emissions at integrity compromised oil and gas wells. In this work package, existing data relevant to CH_4 oxidation in soils at and around oil and gas wells was collated and analyzed with a view to appraising use of the process in natural soils to mitigate GHG emissions at integrity compromised oil and gas wells. Data comprised of soil gas concentration and surface efflux measurements of CH_4 and CO_2 (including stable carbon isotopes where available) associated with 3 recent British Columbia research projects was attained. Collated data was evaluated in the context of characterizing the extents and rates of CH_4 oxidation occurring in natural soils in BC by 2 generalized methods (depending on quality and availability of data for each study) including; a) mass-balance approach, and, b) stable carbon isotope enrichment techniques[15]. Learnings from this WP will provide insight on the potential for natural soils based CH_4 oxidation to mitigate GHG emissions at integrity compromised wells sites, inform additional data collection in WP 2 (i.e., by identifying sites of interest and data gaps) and aid the refinement and calibration of relevant reactive transport models developed in WP3. Data sources attained and methods to evaluate CH_4 oxidation used including findings from this exercise are outlined below.

2. Methods

2.1. Potential CH_4 Oxidation Data Collation

While many studies that report on fugitive gas leakage and soil gas effluxes associated with oil and gas well integrity failure exist, often the data collected is insufficient to robustly characterize CH_4 oxidation or explicit consideration of the process has not been made for various reasons. Here, an exhaustive review of existing studies where data which could be used to understand CH_4 oxidation was attained but not fully utilized was made. Ultimately, three separate studies where suitable quality and extents of data were available to better understand CH_4 oxidation processes were identified and reviewed. A brief description of each study provided as follows.

Study 1: Towards Optimizing Well Plug and Abandonment in British Columbia through Data Analytics, Field Investigations and Predictive Modelling (BC OGRIS Research Project ES-Wells-2021-02)

This investigation formed the precursor and motivation for the current study and focused on evaluating the performance of P&A configurations in BC with respect to well integrity failure. The study showed 5 out of 10 wells examined were potentially suffering minor levels of integrity failure. Moreover, the magnitude of well leakage from the identified integrity compromised wells was generally small compared to other sources (i.e. being significantly less than the emissions associated with a single dairy cow at each well). Most importantly it was shown that a potentially significant portion of released CH_4 was likely being consumed in soils and converted to CO_2. This assertion was made based on the anomalous levels of CO_2 observed emitting from soils at wells identified as potentially suffering integrity failure. However, as the focus of the study was on evaluating P&A configuration performance and general well integrity status (i.e. determining if leakage was occurring or not), the data was not used to fully evaluate the potentially observed CH_4 oxidation processes occurring in the soils. Thus, in this project we re-evaluate this data to better understand rates and extents of CH_4 oxidation occurring by enhanced evaluation of soil CH_4 and CO_2 efflux measurements using a mass balance approach and stable carbon isotope data.

Study 2: Identification, spatial extent and distribution of fugitive gas migration on the well pad scale. O.N. Forde, K.U. Mayer, D. Hunkeler. Science of The Total Environment, Volume 652, 2019, Pages 356-366.

In this study published in 2019, the investigators evaluated 17 well sites in NE BC for signs of potential integrity failure, again focused mostly on whether signs of leakage were apparent and not explicitly evaluating for the presence of, or extents, of CH_4 oxidation that might be occurring. However, significant data on CH_4 and CO_2 soil gas fluxes and stable carbon isotope ratios of observed gases were attained which could be re-evaluated to identify and characterize any natural soils based CH_4 oxidation occurring. Here, the authors of this study were contacted and raw data from the study attained and re-evaluated through enhanced evaluation of soil CH_4 and CO_2 effluxes in terms of mass balance and by consideration of stable carbon isotope data.

Study 3: Hudson's Hope Unsaturated Zone Controlled Natural Gas Release Experiment

This study, conducted in 2017 and reported in several peer-reviewed journal articles[8,16], involved simulating subsurface wellbore leakage by injecting natural gas (a total of 30 m^3) into thick unsaturated deposits at 12 m depth in a region of petroleum development in Western Canada. The response was monitored by measuring depth discrete CH_4 and CO_2 concentrations and emissions on the ground surface. The primary focus of the work was to evaluate the extent and patterns of subsurface gas migration and magnitudes of emissions to atmosphere. Consequently, little focus was placed on identifying and evaluating the presence and nature of CH_4 oxidation in the soils during and after the experiment, despite the potential for data relevant to this process being collected. Here, the authors were contacted and raw data from the study attained and re-evaluated to determine the presence and nature of CH_4 oxidation through enhanced evaluation of effluxes measured in terms of mass balance and by consideration of stable carbon isotope data.

2.2. CH_4 Oxidation Data and Evaluation Approaches

In order to evaluate the occurrence and extent of natural soils based CH_4 oxidation, it is necessary to have suitable data that can indicate the removal and conversion of CH_4 to CO_2 over appropriate spatial and/or temporal scales through the soils. Such data includes the influx/leakage rate of CH_4 into soils, understanding on the timescales, volumes and distances through soils within which CH_4 migration/oxidation occurs and measurement of concentrations and fluxes of both CH_4 and CO_2 at discrete points throughout the soil volume/travel path (including the fluxes at the soil/atmosphere interface). In addition, data on stable carbon isotopes of both CH_4 and CO_2, ancillary weather parameters (e.g. air temperature, barometric pressure, wind speed and rainfall) and soil conditions (e.g., moisture content and permeability) are needed to further contextualize, estimate and evaluate rates and extents of any CH_4 oxidation observed. The scope for characterization of CH_4 oxidation and the methods that can be used to evaluate it depend on the exact measurements made and the nature of data available. Assuming some or all of these data are attained or available; two methods can potentially be used to evaluate extents and rates of CH_4 oxidation occurring in soils, including; a) a mass balance approach, and, b) a stable carbon isotope approach (Figure 1).

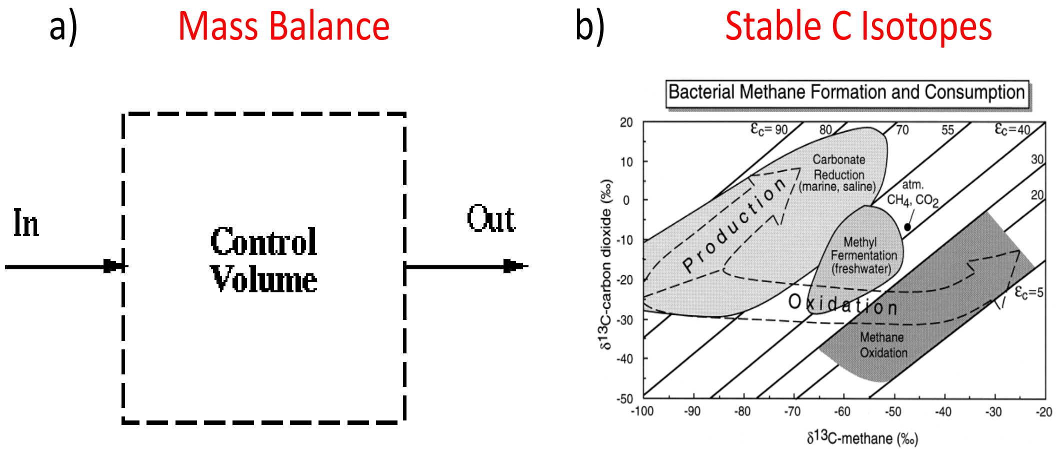


Figure 1: A conceptual depiction of two generalized approaches that can be used to determine the extent and rates of CH_4 oxidation in soils depending on the data available.

For the mass balance approach, the aim is to quantify inputs and outputs of CH_4 in the soil system to determine its overall balance. In a general sense, this involves understanding the methane source (e.g., well leakage rate) and sinks (e.g., microbial oxidation) through measuring the CH_4 and CO_2 fluxes entering, travelling through and leaving the soil. From these data (if correctly and suitably measured) and understanding the spatiotemporal context of the system, it is then possible to estimate CH_4 oxidation extents in the soil through consideration of the fugitive CH_4 mass balance to determine net depletion of CH_4. The mass balance approach works based on the principle of conservation of mass. It assumes that the total amount of methane entering a soil system from a leaking well must be equal to the total amount leaving through processes like diffusion, transport, and microbial oxidation. By quantifying these inputs and outputs, the approach provides insights into the overall methane balance in the soil system. If microbial methane oxidation is significant, it acts as a sink, reducing the net emissions from the soil to the atmosphere while simultaneously increasing CO_2. From a mass balance approach, the extent of CH_4 oxidation can generally be determined using the following equation:

$$f_{oxm} = 1 - \frac{J_{out}}{J_{in}}$$

Where f_{oxm} represents the fraction (as a ratio or %) of methane that has been oxidized (i.e. removed) from the system while $J_{in/out}$ represent the fluxes of methane into and out of the system respectively.

The stable carbon isotope approach involves analysing the carbon isotopic composition of CH_4 and CO_2 molecules in the soil system to differentiate sources and assess CH_4 oxidation and CO_2 generation rates. The approach involves measuring isotopic fractionation shifts of the carbons atoms that form the CH_4 and CO_2 molecules, which are determined by the occurrence and extents of methane oxidation in the soil system. The approach is based on the principle that there are distinct isotopic signatures of CH_4 from different sources and that such source CH_4 is then subject to fractionation of isotopes during microbial CH_4 oxidation. When methane is oxidized by microbes, lighter isotopes (i.e. ^{12}C) are preferentially consumed, leading to changes in the isotopic ratio of remaining CH_4 and resulting CO_2. By analysing isotopic changes between a source and oxidized gas, an estimate of the oxidation fraction (i.e. the extent to which oxidation has likely occurred) can be attained according to the following equation.

$$f_{oxio} = \frac{\delta_{out} - \delta_{in}}{1000(\alpha_{ox} - \alpha_{trans})}$$

Where f_{oxio} is the fraction of methane oxidized, $\delta_{in/out}$ represents the stable carbon isotope ratios of the source gas and end member and $\alpha_{ox/trans}$ represents an enrichment factor.

Both methods works by leveraging different principles and techniques to assess methane dynamics in soils associated with a leaking oil and gas well. The mass balance approach quantifies overall methane fluxes while the stable carbon isotope approach provides insights into methane sources and oxidation rates. When possible, combining these methods provides a more comprehensive understanding of methane dynamics and the effectiveness of microbial CH_4 oxidation in natural soils around a leaking oil or gas well. For the studies identified in 2.1, we now apply, where possible, the described methods in order to characterise and evaluate the extents and rates of CH_4 oxidation occurring.

3. Results and Discussion

3.1.Data from BC OGRIS Research Project ES-Wells-2021-02

Data from the five well sites identified in BC OGRIS Research Project ES-Wells-2021-02 as potentially exhibiting integrity failure was further evaluated in the current project with respect to better understanding the extents of CH_4 oxidation. This was achieved through a mass balance and stable carbon isotope approach whereby overall total site wide excess emissions for both CH_4 and CO_2 were derived from measured flux footprint contours as shown in Figure 2 (interpolated by kriging and determining excess emissions against site background measurements). Here, the presumed extent of CH_4 oxidation potentially occurring (i.e., based on all excess GHG emissions observed at each site assumed as derived from well leakage with excess CO_2 generated only from CH_4 oxidation) and the total reductions in GHG emissions afforded by soil-based CH_4 oxidation (i.e., estimating the total GHG emissions in CO_2 equivalents if no CH_4 biodegradation was occurring in the surficial soils) were calculated based on a simple mass balance approach.

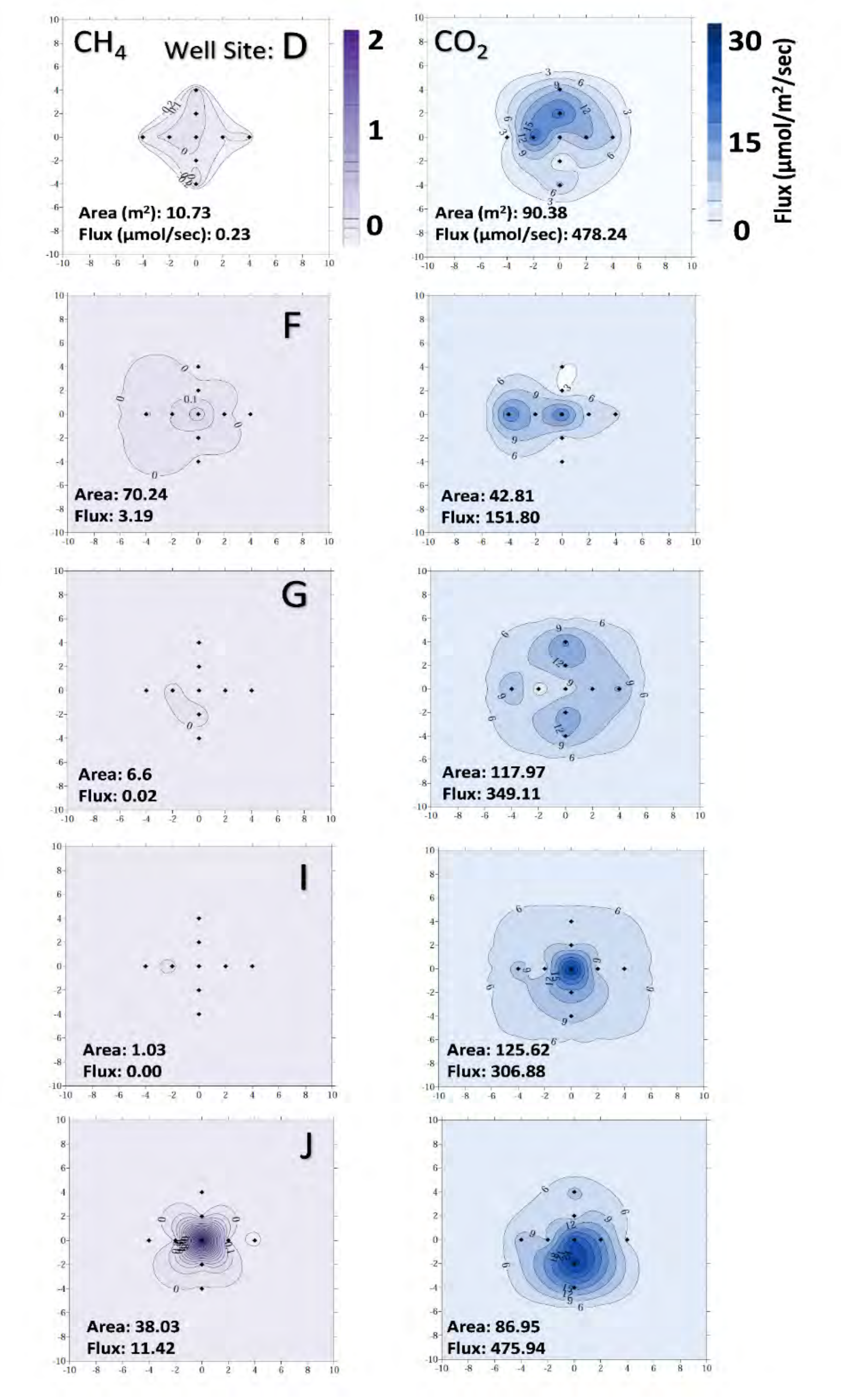


Figure 2: Plan view contour plots (interpolated by kriging) of CH_4 and CO_2 efflux in µmol/m^2/sec at and around the examined wellheads (i.e. located at 0, 0) for sites exhibiting excess CH_4 and/or CO_2

fluxes. Also indicated in bottom left of each pane is the overall excess flux footprint area and total excess flux in µmol/second (relative to site specific background measurements)

An example of this approach is shown in Figure 3 for wellsite WA22800. Here, excess levels (i.e. levels above observed background ranges) of CH_4 and CO_2 are calculated as a 3-D surface (i.e. with the volume under the curve above background levels assumed excess emissions associated with leakage) and combined to determine overall site wide emissions (i.e. in CO_2 equivalents, assuming 1 kg of CH_4 equates to 80kg of CO_2 over a 20-year period). Results are summarized in Table 1.

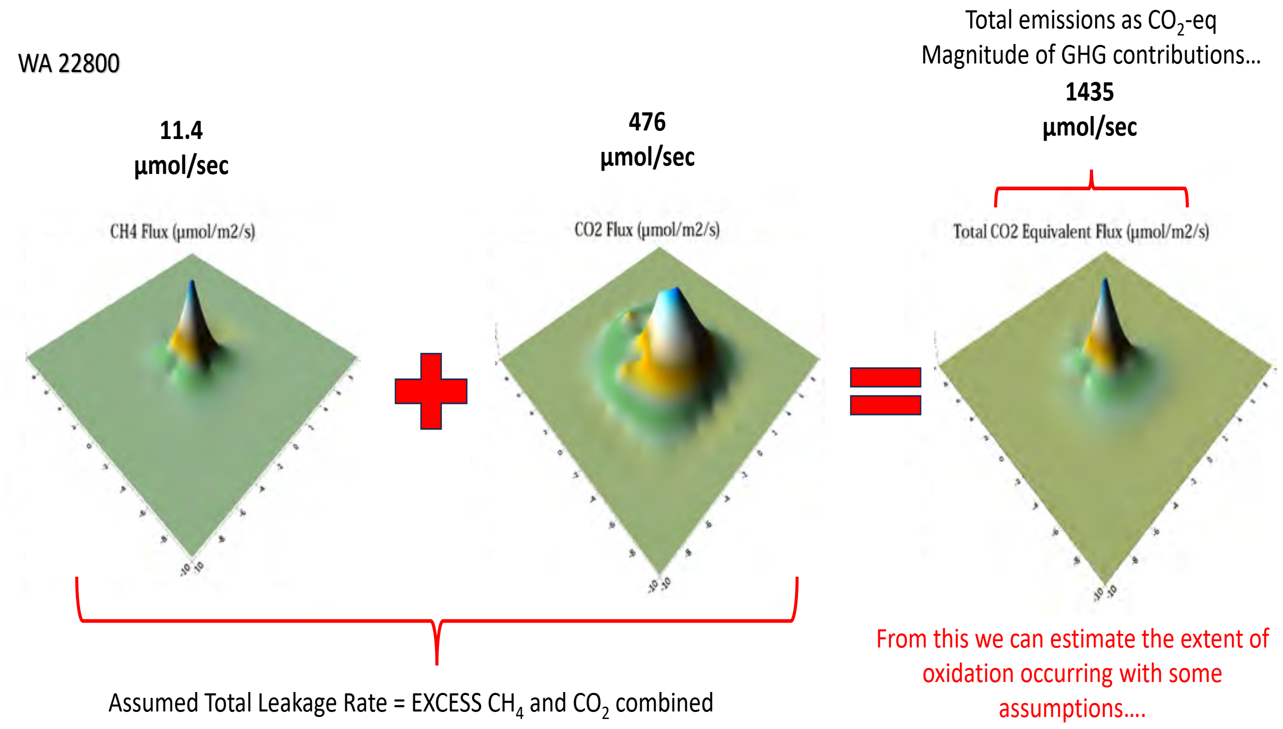


Figure 3: Conceptual overview of the mass balance approach taken to determine the extent of CH_4 oxidation potentially occurring in natural soils at well sites examined in BC as part of Project ES-Wells-2021-02. In the approach, excess CH_4 and CO_2 emissions are determined for each site from a 3D flux footprint curve (interpolated by Krigging and determining the area under the curve as total flux) and combined to estimate total site wide excess emissions in CO_2 equivalents (including both CH_4 and CO_2). From this estimated total, the extent of degradation can be inferred by assuming all excess CO_2 (i.e. in excess of background observations) comes from CH_4 oxidation.

Table 1: Summary of overall total site wide excess emissions, the presumed extent of biodegradation potentially occurring and the total reductions in GHG emissions afforded by soil-based CH_4 oxidation.

Well Site	Excess CH_4 flux (µmol/sec)	Excess CO_2 flux (µmol/sec)	Total Excess Emissions in CO_2-eq (µmol/sec)	Total Excess Emissions in CO_2-eq (kg/yr)	Potential Extent of CH_4 Degradation (%)	Potential Maximum CO_2-eq Emissions with no CH_4 degradation (kg/yr)	Degradation Based Reduction in CO_2-eq (%)
D	0.23	478.2	497.4	690.2	~100	55,743	98.8
F	3.19	151.8	420.0	582.8	~98	17,698	96.7
G	0.02	349.1	350.9	486.9	~100	40,691	98.8
I	0.00	306.9	307.0	426.0	~100	35,769	98.8
J	11.42	475.9	1,434.9	1,991.0	~98	55,489	96.4

Next, in order to estimate potential oxidation rates for CH_4 in natural soils structures based on field observations an additional simple mass balance style calculation was performed assuming that all excess CO_2 observed is derived from CH_4 oxidation. Furthermore, it is assumed that oxidation must be occurring through and across an average area of 100 m^2 of soils (a relevant average footprint size as seen from figure 2). Based on this an estimated leakage rate and mass of CH_4 oxidized per day can be derived, followed by an estimated oxidation rate with results for the five sites examined shown in Table 2.

Table 2: Summary of overall estimated site wide leakage rates, mass of CH_4 oxidized and resultant estimated oxidation rate based on an assumed area of oxidation of $100m^2$.

Well Site	Estimated Leakage Rate (kg/CH_4/day)	Estimated Mass of CH_4 Oxidized (kg/CH_4/day)	Estimated CH_4 Oxidation Rate (g/m^2/day)
D	**0.69**	**0.66**	**6.61**
F	**0.58**	**0.21**	**2.10**
G	**0.49**	**0.48**	**4.83**
I	**0.42**	**0.42**	**4.24**
J	**1.98**	**0.66**	**6.58**
AVERAGE	**0.83**	**0.49**	**4.87**

Finally, stable carbon isotope data from this study were further evaluated (shown in Figure 4) to explore potential extents of CH_4 oxidation. Here, detected methane at wellsite WA22800 was seen to be enriched in ^{13}C compared to what might be expected for typical thermogenic gas signatures (i.e. at -33.7 ‰ compared to typical values of between -40 to -50 ‰ giving rise to a potential shift in $\delta^{13}C$-

CH_4 of 7 to 17 ‰). Meanwhile emitting CO_2 at the site was seen to be depleted in ^{13}C (i.e. with a more negative $\delta^{13}C$-CO_2) compared to background locations (at -30 ‰ compared to -17 ‰ in the case of wellsite J for a potential shift in $\delta^{13}C$-CO_2 of approximately -13 ‰). In this case, the shifts in $\delta^{13}C$ appear consistent (i.e. 10 – 15 ‰) supporting that the CO_2 emitting as described above is likely to be derived from natural soils based oxidation of fugitive CH_4. In addition, the fraction of CO_2 generated (i.e. the inverse of methane oxidized) was determined using the open system stable carbon isotope technique (as described in section 2.2). Here a background $\delta^{13}C$-CO_2 value as observed from field samples of -18 ‰ was used as the input value in conjunction with the most ^{12}C-depleted signature for wellhead soil gas at wellsite WA22800 of -30 ‰. Consideration of these $\delta^{13}C$-CO_2 values indicates a potential estimate for fraction of observed CO_2 generated by CH_4 oxidation of ~48%.

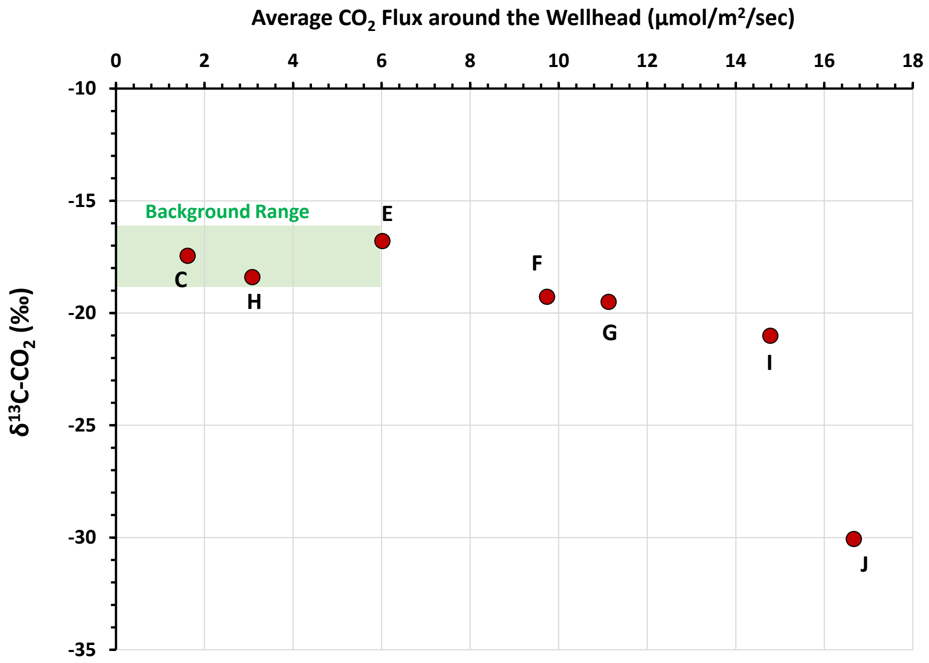


Figure 4: Cross plot of $\delta^{13}C$-CO_2 versus average flux of CO2 around the wellhead by site examined showing some sites exhibiting depletion in ^{13}C (i.e. with a more negative $\delta^{13}C$-CO_2) compared to background locations.

3.2.Data from Forde *et al.*

In the study conducted by Forde *et al.* 15 out of 17 wells examined were concluded to be showing signs of integrity failure with fugitive gas efflux at the well pad surface. Here, raw data including CO_2 and CH_4 flux measurements and stable carbon isotope data from field surveys were attained from the authors and reviewed to better understand the presence and extent of any potential soils based CH_4 oxidation occurring at the examined well sites visited. In particular, CO_2 and CH_4 flux data for each site (individual point values and site wide averages) were evaluated against stated background fluxes and other reported values associated with leakage in the literature. The aim was to verify/reconfirm which sites were potentially exhibiting integrity failure and fugitive gas release and further identify which sites exhibited signs of soils based CH_4 oxidation occurring through either the presence of high levels of CH_4 and CO_2 co-emission and/or particularly high levels of CO_2 emissions. Wells were assigned as showing either low, moderate or high potential for CH_4 oxidation in soils and re-classified as potentially leaking, potentially not leaking and leaking with certainty by considering a combination of point and average concentrations and fluxes of GHGs (with results in Table 3). Meanwhile the reported maximum fluxes of CH_4 and CO_2 for each site were cross-plotted and are shown in Figure 5 with typical baseline ranges for both delineated by dashed lines and a shaded green area.

Table 3: Re-evaluation of Forde *et al.* data from 17 well pads in BC indicating levels of CH_4 oxidation potentially occurring and a new leakage assessment compared to that made by Forde *et al.*

Well Site	Max CH_4 flux	Site Ave. CH_4 Flux	BG CO_2 Flux	Site Ave. CO_2 Flux	Max CO_2 flux	Potential Signs of CH4 oxidation	Forde *et al.* leakage Assessment	New leakage Assessment
1	4.2	n/a	1.6	2.4	7.7	Low	🟧	🟧
2	4.8	0.54	5.7	2.9	9.1	Low	🟧	🟧
3	1.4	0.07	2.1	3.1	16	Moderate	🟧	🟧
4	0.065	0.008	1.1	2.6	18	Moderate	🟧	🟧
5	1.3	0.08	0.63	3.2	20	Moderate	🟧	🟧
6	0.31	0.01	1.6	2.7	9.3	Low	🟧	🟩
7	0.44	0.004	1.7	3.9	10	Low	🟧	🟩
8	0.083	0.006	3.4	6.4	27	High	🟧	🟧
9	0.34	0.02	1.1	1.9	3.7	Low	🟧	🟩
10	7.5	0.44	0.89	1.2	7.3	Low	🟧	🟩
11	3.9	0.04	0.79	0.8	8.1	Low	🟧	🟩
12	180	6.8	0.94	1.8	10	Low	🟧	🟨
13	0	-0.002	3.8	2.8	7.1	Low	🟩	🟩
14	0.025	0.001	3.1	12	32	High	🟧	🟧
15	0	0.001	6.7	6.4	19	Moderate	🟩	🟧
16	0.12	0.006	2.2	3.2	9.6	Low	🟧	🟩
17	2.2	0.22	0.71	1.9	5.9	Low	🟧	🟩

Well Status Assignment based on CH_4 and CO_2

🟧	🟩	🟨
Potentially Leaking	Potentially NOT Leaking	Leaking with Certainty

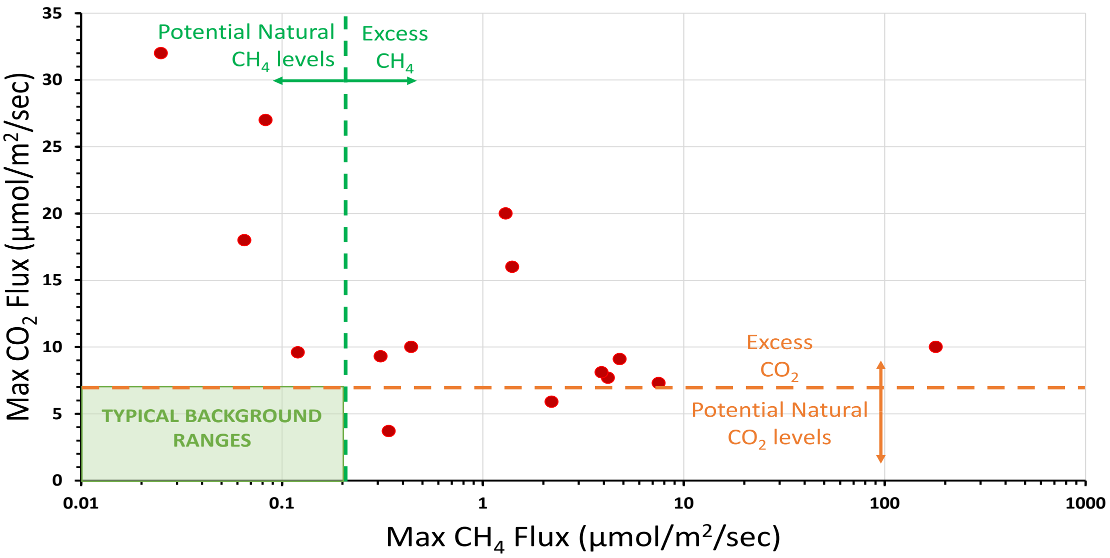


Figure 5: Cross plot of maximum CH_4 and CO_2 fluxes measured at each well site examined in Forde *et al.* in comparison to typical background ranges showing where observations were potentially anomalous and where soils based CH_4 oxidation may be occurring even if no excess CH_4 was detected.

From this new review of reported data in Forde *et al.*, well sites 8, 14 and 15 were identified as showing the greatest signs of potential soils based CH_4 oxidation (with the most anomalously high CO_2 fluxes being evident) and selected for further data evaluation. Consequently, all individual CH_4 and CO_2 fluxes measured at each of these sites were cross- plotted to show the extent of anomalous CO_2 present compared to background levels (Figure 6). Additionally a mass balance 3-D surface approach as described in section 3.1 was taken to evaluate the extent of excess CO_2 and derive the potential leakage magnitudes and resultant oxidation rates with results shown in Figure 7 and Table 4.

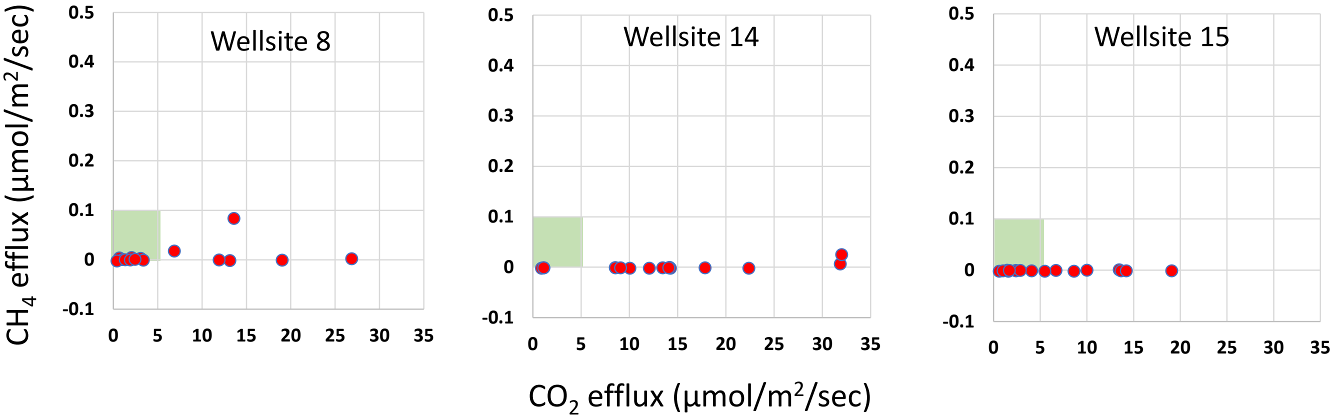
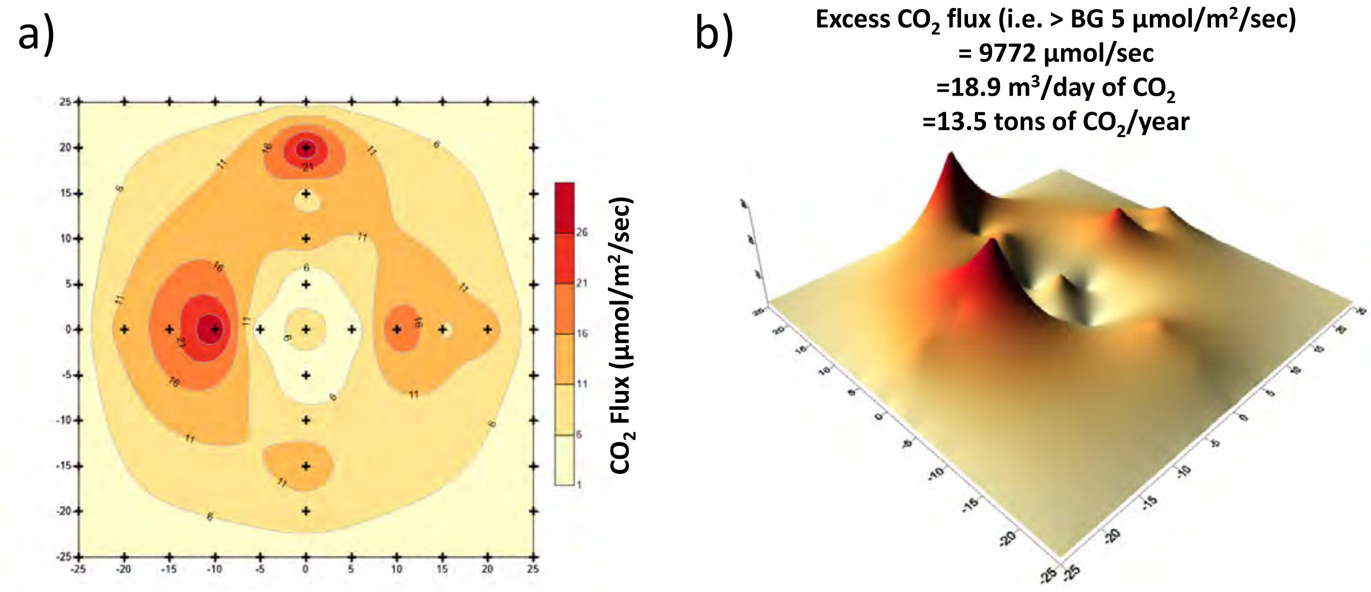


Figure 6: Cross-plots of CH_4 and CO_2 efflux for sites identified in Forde *et al.* where potential for significant oxidation was identified showing very high levels of CO_2 flux despite low levels of CH_4 emissions. The green shaded square represents observed background ranges of fluxes, showing many measurements were outside of these ranges and therefore appear anomalous.



Excess CO$_2$ flux (i.e. > BG 5 µmol/m^2/sec)
= 9772 µmol/sec
=18.9 m^3/day of CO$_2$
=13.5 tons of CO$_2$/year

Figure 7: An example of an emissions contour and 3D surface plot for site S14 showing excess CO$_2$ emissions around the wellhead (as high as 30 µmol/m^2/sec at multiple locations), far greater than background conditions observed (i.e. 3.1 µmol/m^2/sec)

Table 4: Estimated parameters from Forde *et al.* data for sites identified to be potentially exhibiting soils based CH$_4$ oxidation according to anomalous levels of CO$_2$ efflux compared to background levels. Values were derived from contour plots (interpolated by Krigging and determining the area under the curve as total flux) with volumes one standard deviation above background considered excess emissions (with 5 µmol/m^2/sec assigned as background value for site S8 and S14 and 7 µmol/m^2/sec set as baseline for site S15).

Site	Site total flux kg CO$_2$/yr above BG	Site total excess flux (m^3/day @STP)	Estimated leakage rate in kg CH$_4$/day	Estimated CH$_4$ leakage rate in kg/yr	CO$_2$ eq with no degradation in kg/yr	Estimated CH$_4$ oxidation rate (g/m^2/day) assuming 100m^2 reactive soil zone
S8	3,251	4.5	3.2	1,182	99,306	32.4
S14	14,437	20.1	14.4	5,250	441,009	143.8
S15	3,041	4.2	3.0	1,106	92,906	30.3

Finally, isotope data attained during these investigations was reviewed and analysed to determine the potential fraction of CH$_4$ oxidized using the open system stable carbon isotope technique (as described in section 2.2). Here, the fraction of CH$_4$ oxidized was calculated using the average observed SCVF and soil gas based δ^{13}C-CH$_4$ signatures (termed the average case) and the potential maximum δ^{13}C-

CH_4 with the largest differential observed between site SCVF and soil $\delta^{13}C$-CH_4 detected. An α value of 1.025 was assumed at reported by Forde *et al.* The results, shown in Table 5, suggest that on average ~12% of detected CH_4 was oxidized while a potentially maximal extent of ~52% oxidation was potentially observed at one of the well sites visited.

Table 5: Observed $\delta^{13}C$ values and resultant fraction oxidized calculated for the average and maximal cases observed using the Forde *et al.* data.

Parameters	Average	Maximum
δ OUT (‰)	-43.9	-37
δ IN (‰)	-47	-50
Fraction of CH_4 oxidized (%)	12.4	52

3.3. Data from Hudson’s Hope Unsaturated Zone Experiment

Data from this experimental study was previously reported in two journal articles where little focus was placed on identifying and quantifying soils based CH_4 oxidation. A key finding from this study, however, was that barometric pumping was a very important factor for controlling the movement of gas from a subsurface leakage point (i.e. 12 m depth in this experimental configuration) to the surface. In this case, decreasing pressure acts to “pull” gas out of the subsurface into the atmosphere faster, while increasing pressure acts to “hold” fugitive gas in the ground for longer. Here, the raw data from this study was attained and reviewed to evaluate whether further quantification on extents and rates of CH_4 oxidation could be achieved by one of the two generalized approaches as outlined in Section 2.2. Regrettably, upon review and consideration of the data it became clear that the barometric pumping phenomena identified renders the mass balance method for quantifying CH_4 oxidation unusable. Here the physical process of gases being “pulled out” of, or “held within”, the ground due to barometric pumping means that meaningfully evaluating oxidation through mass balance is not possible. The principle concept being that if physical processes such as barometric pumping significantly influence or control the amounts of gases coming into or out of the ground surface, it is difficult to attribute any observations to oxidation alone. Meanwhile, barometric pumping will also alter the reactive transport time/duration by either expediting or retarding CH_4 as it moves towards surface. In the case of increased residence time in the subsurface (i.e. during high-pressure periods that hold gases in the soils), more oxidation can potentially occur while for expedited gas transport (i.e. during low-pressure periods where gas is pulled out of the soils) less oxidation can occur. While somewhat disappointing that this data could not be used further, this issue raises a very important point that will affect the efficiency and extent of any natural soils based CH_4 oxidation associated with oil and gas wells. Here, it is clear processes like barometric pumping can affect the extents and rates of CH_4 oxidation within soils and clearly influence the potential use of soils based CH_4 oxidation in any management or mitigation strategy for integrity-compromised wells and thus may need to be managed. A diagram showing the process of barometric pumping as identified during the experiment is shown in Figure 8.

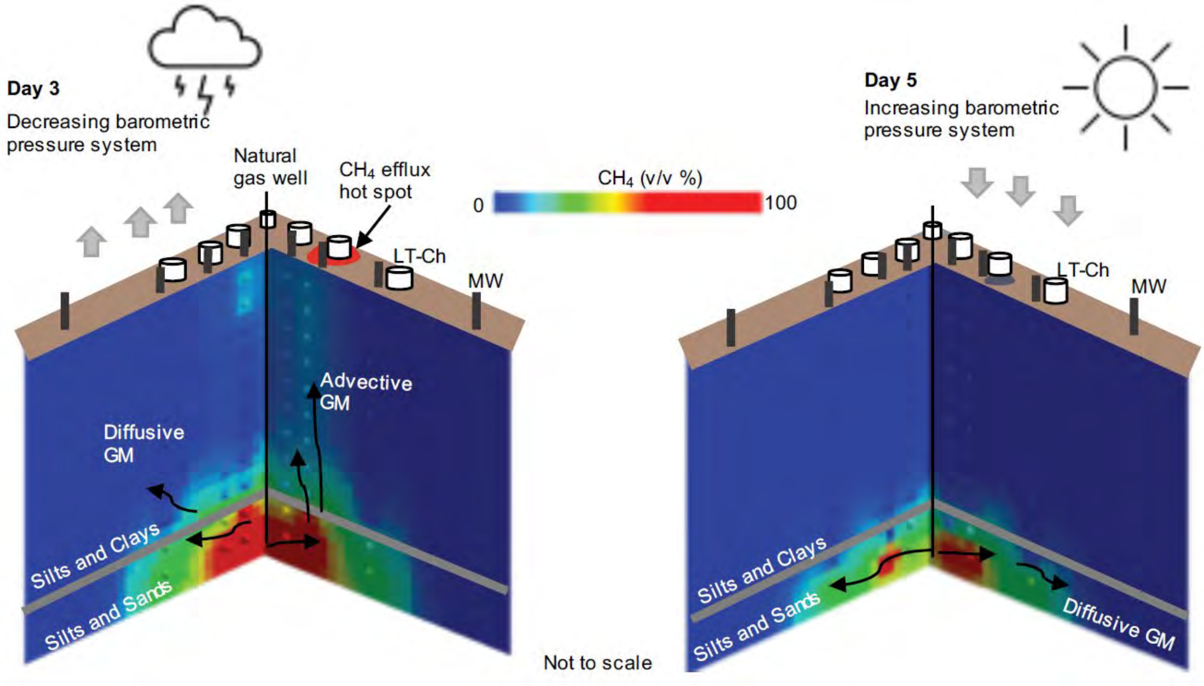


Figure 8: Diagrammatical figure showing barometric pumping of fugitive gas as observed during the Hudson's Hope Unsaturated Zone Injection experiment, whereby: a) during decreasing pressure gas is pulled towards ground surface and emits to atmosphere shortening reactive residence time in the soils, and, b) increasing barometric pressure whereby fugitive gas is held in the ground increasing reactive residence time. This physical effect makes it difficult to use the associated data to estimate oxidation rate and extents.

While the mass balance approach was not of use with these data, the stable carbon isotope data was potentially useable and was further evaluated by the oxidation extent method as described in section 2.2. Here extents of CH_4 oxidation (i.e. as fraction of CH_4 oxidized) were estimated based on the $\delta^{13}C$-CH_4 values measured during the experiment; including on day 35 and day 400 post gas injection with the results shown in Table 6.

Table 6: Observed average and maximum $\delta^{13}C\text{-}CH_4$ values and resultant extent of oxidation estimates on day 35 and 400 during the Hudsons Hope Unsaturated Zone Injection Experiment.

Parameters	35 Day Average	35 Day Maximum	400 Day Average	400 Day Maximum
δ OUT (‰)	-29.6	-25.6	-25.1	-18.6
δ IN (‰)	-40	-40	-40	-40
Fraction of CH_4 oxidized (%)	41.6	57.6	59.6	85.6

4. Discussion and Conclusions

This work package sought to use existing data from studies where quantifying CH_4 oxidation was not a priority, yet data of potential use to do so was collected. These data were attained and re-evaluated through the lens of identifying and quantifying soils based fugitive CH_4 oxidation. The following form the main findings from this exercise:

- Signs of CH_4 oxidation in natural soils were consistently detected (at least at some level) for all cases we examined (including at real wells and during the experimental controlled release investigation) demonstrating the process is ubiquitous, robust and adaptable. This is more so considering the cases examined spanned different soil and leakage types over a range of environmental conditions inferring that soils based CH_4 oxidation can proceed relatively favorably under a range of conditions in British Columbia and is not a rare occurrence.
- In terms of extents of oxidation (i.e. the fraction of released CH_4 being consumed), the two main approaches employed (i.e. mass balance and stable carbon isotope analyses) generated slightly differing estimates, while different extents were also seen between sites/studies. Regardless of differences, these analyses show natural soils have significant assimilation capacity for CH_4 through oxidation having propensity to degrade >50% (often considerably more) of any fugitive gas in the cases that we examined.
- Oxidation rates were estimated using data from the five sites examined in BC OGRIS Research Project ES-Wells-2021-02 and three select sites examined in Forde *et al.* using a generalized mass balance approach. Oxidation rates ranged from as low as 2 g CH_4/m^2 of soils/day to as high as 144 g CH_4/m^2 of soils/day, with an average ~30 g CH_4/m^2/day.
- Critically, it can be seen from these estimates that CH_4 oxidation rate is strongly controlled by the magnitude of leakage. That is, a greater mass of CH_4 being released into the soils leads to more being oxidized and a higher oxidation rate. This finding is in line with other studies

around landfill cover oxidation rates[15] and infers that natural soils might typically have a capacity to oxidize more CH_4 than is released (at least for the wells examined here).

- Overall, the CH_4 oxidation rates we report here are not potential maximums, but conservative, observed rates and perhaps demonstrate rates at which BC soils can comfortably assimilate CH_4. Higher rates are almost certainly achievable with optimized conditions and larger leakage rates; however more work is needed to determine potential theoretical maxima.

WP2: Field Examination of CH_4 Oxidation in Natural Soils of BC

1. Introduction and Background

To date, no purpose-specific field investigations have collected data from which the extents, rates and controlling parameters associated with CH_4 oxidation in natural soils at integrity-compromised wells can be robustly characterized. Here, to address this data and knowledge gap, a new and uniquely designed 2 week field investigation focusing on 4 potentially integrity compromised wells identified in Project ES-Wells-2021-02 was undertaken in August 2023. The overall aim of the field campaign was to better understand CH_4 oxidation in natural soils of BC at the well sites of interest and attain measurements and data that could allow the estimation of oxidation extents and rates as well as identify the microbial communities that may be performing the oxidation process. With this in mind, a more detailed monitoring/sampling program than previously undertaken in Project ES-Wells-2021-02 was enacted, alongside more focused data analyses associated with four of the integrity compromised well sites where CH_4 oxidation in natural soils was previously qualitatively identified as occurring. A map showing the location of the four sites examined is provided in Figure 1, while Table 1 provides a summary of previously measured soil gas fluxes at these sites that were identified as likely to be suffering integrity failure. Figure 2 shows wellsite WA22800, where the greatest signs of leakage and CH_4 oxidation in soils was detected, which formed a key location for these new investigations. Herein, methods employed in these new investigations are described before results are presented and discussed in terms of rates and extents of CH_4 oxidation observed occurring in soils at the examined sites and microbes detected that may be involved.

Table 1: Summary of wellsites examined in new focused field investigations including previously observed fluxes of CH_4 and CO_2 from Project ES-Wells-2021-02.

Wellsite (WA)	**No. of Flux Measurements Previously Taken**	**Well Head Area Fluxes (µmol/m²/sec)**			**Background (µmol/m²/sec)**	
		Ave. CH_4	**Max. CH_4**	**Ave. CO_2**	**Ave. CH_4**	**Ave. CO_2**
9876	11	0.006	0.108	10.93	-1.14	0.519
12660	13	-0.009	0.012	11.13	-0.032	5.29
21083	12	-0.017	0.005	14.78	-0.004	5.87
22800	13	0.73	3.01	16.67	-0.03	3.96

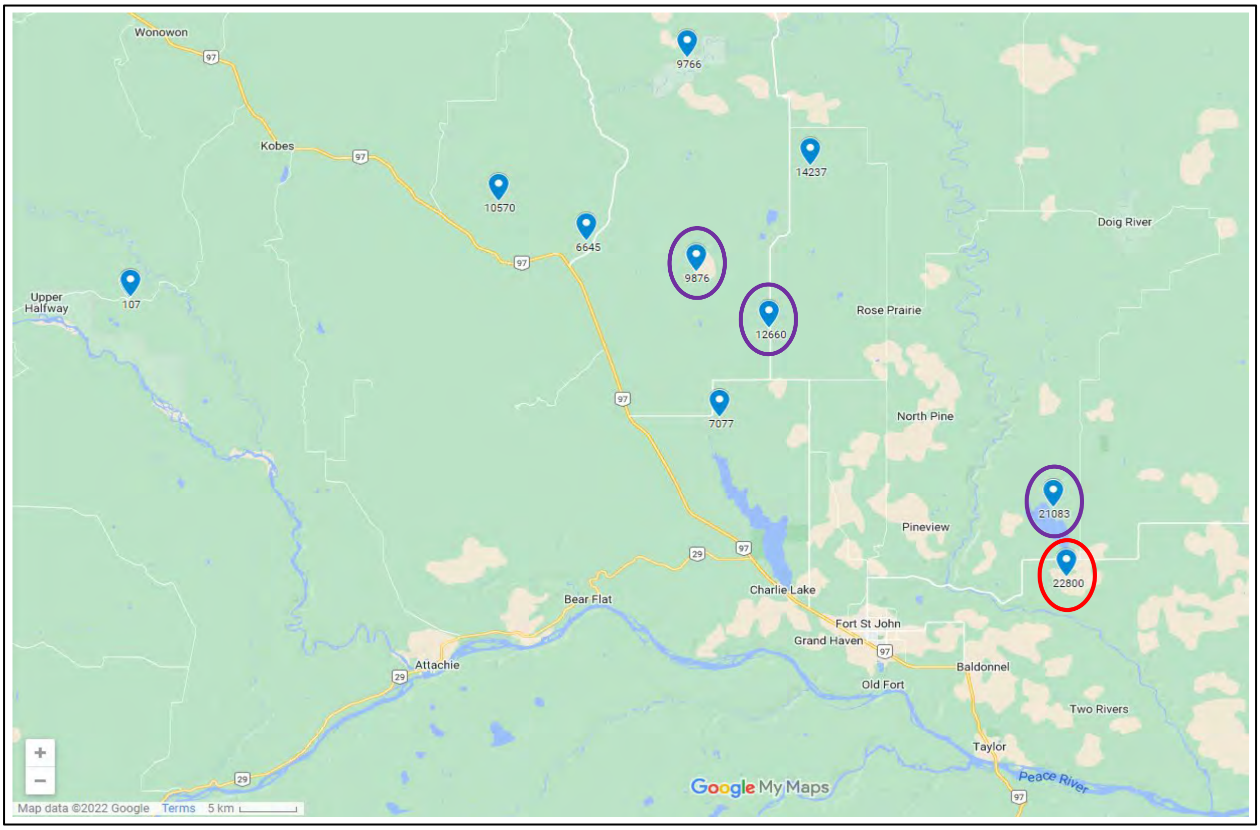


Figure 1: Map showing location of wells previously examined in the Fort St John area during Project ES-Wells-2021-02 (blue pinpoints) with wells further examined in the current project circled (with the red circle identifying the site with greatest signs of leakage and CH_4 oxidation in soils).

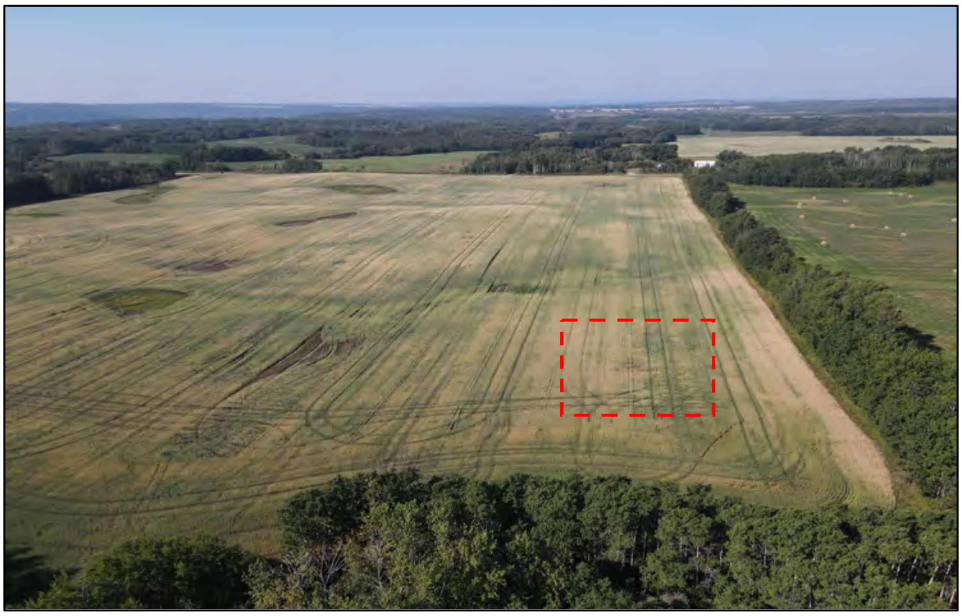


Figure 2: Aerial view of wellsite WA22800 identified in August 2022 with potential integrity failure and at which CH_4 and CO_2 were observed co-emitting to atmosphere, indicating CH_4 oxidation in natural soils around the well. This site formed a key focus of these new investigations.

2. Methods

2.1. Field Investigations

Field investigations at the four well sites took place during the last week of August and first week of September 2023 and included a range of monitoring and measurement methods. During field investigations, candidate wellheads were first re-located according to regulatory held coordinates using a GPS (accuracy to +/- 2m) and visual identification. After locating the well head a sampling grid generally formed of 9 points in a cross hair pattern (oriented North with each sampling point 2 m apart) was established, centered on the assumed well head location (Figure 3). A dynamic flux chamber system was then used to measure CH_4 and CO_2 efflux from the soils at each sample point above and around the examined P&A'd well in spatial survey mode. The flux chamber was comprised of an Eosense® eosAC Multi-Species Soil Flux Chamber coupled with a tuneable diode CH_4 and CO_2 laser and Campbell Scientific CR1000 data logger. The chamber was used in two general configurations, including; a) survey mode (attaining spatial data around the wellhead at a given time), and, b) long term continuous mode (i.e. measuring continuously at a single point over some hours). Moreover, both of these configurations were also deployed first at surface (termed surficial measurements) and then over the top of 1m deep auger holes (termed sub-soil measurements). During spatial surveys, the chamber was moved between the different sample points at a site to attain single, short duration (i.e. 5 minute) flux measurements. For **surficial** flux measurements, prior to flux chamber deployment, a hand auger (12 cm in diameter) was used to penetrate the topsoil and root zone (i.e. to around 10 cm depth) at each measurement location and a cylindrical collar positioned around the shallow hole (i.e. pushed 5 – 10 cm into the soils) to create a more effective seal. For **sub-soil measurements**, an 8" hole was advanced to 1 m depth below ground surface using an electric auger drill with the cylindrical collar pushed into the auger hole opening, allowing gases emitting from 1 m depth to be measured. Regardless of surface or sub-soil configurations, the dynamic flux chamber was then deployed on the cylindrical collar and operated in manual mode, opening and closing upon initiation, while continuously measuring chamber air CH_4 and CO_2 concentrations (Figure 4 and 5). To yield a flux value (in mass/area/time), a regression line was derived from any rate of change in CH_4 or CO_2 concentration observed during chamber closure and combined with the ideal gas volume (derived from ambient temperature and pressure), the chamber volume ($0.0021m^3$) and cross-sectional area ($0.0182m^2$). In addition to flux measurements at and around the wellhead location, a background location was also monitored at each wellsite. Background locations were established at least 50 m from the wellhead, ensuring ground and vegetation cover remained representative.

Physical soil-gas samples for laboratory analyses were collected at select locations (i.e. typically at the monitoring grid centre point) using the flux chamber systems peristaltic pump and a 25 ml gas-tight syringe system (SGE 25MDR, Restek Thames, England) with Luer Lock Valve. Here, the 2" syringe needle was placed into the flowing soil gas effluent line, purged several times before a sample was taken and transferred into a 12 ml pre-evacuated sampling tube. In addition to soil gas samples, several atmospheric air control samples were also taken for comparative purposes. Samples were stored upside down and analysed for gas composition (CH_4, CO_2, N_2, O_2, and Ar) and stable-carbon isotope ratios (^{13}C/ ^{12}C) of CH_4 and CO_2 in the Isotope Science Laboratory at the University of Calgary (Alberta, Canada) using a Scion 450/456 gas chromatograph (GC). The lower detection limit for hydrocarbon gases is 1 ppm and for non-hydrocarbon gases 50 ppm. Analytical precision and accuracy for gas composition analysis was ±2.5% of the reported concentrations. Stable carbon isotope ratios $^{13}C/^{12}C$ of CH_4 and CO_2 were also quantified at the ISL using continuous-flow isotope-ratio mass spectrometry. Isotopes were measured using a Thermo Trace GC – GC-IsoLink system interfaced to a Thermo Scientific MAT 253 mass spectrometer via a Thermo Conflo IV. The final isotopic ratio results are expressed as δ values using per mil notation relative to the international V-PDB and V-SMOW standards for ^{13}C with an associated accuracy of ±0.5‰.

Select surficial sediment samples were attained from discrete depths (using the electric auger, double bagged in a soil sediment sample bag and stored in a cooler) for 16S rRNA analysis, a powerful technique used to identify and study microbes in various environments, including soils. Here, every microbe has a unique sequence in a specific part of its genetic material called the 16S ribosomal RNA (rRNA) gene, which acts like a fingerprint that can help identify it. The soil samples collected will be used to extract DNA from all the microbes present in the soils, which contains the 16S rRNA genes. The extracted DNA will then be sequenced to determine the order of nucleotides in the 16S rRNA genes, allowing identification of the microbes present in different soils by comparing the sequences to a database of known 16S rRNA sequences. By analyzing soils collected at the suspected integrity compromised wellheads, we can determine which microbes are present and potentially actively involved in methane oxidation, a process where certain microbes consume methane as an energy source.

Finally, relevant local climatic factors were monitored during field investigations (i.e. air temperature, barometric pressure, wind speed and direction, soil moisture, soil temperature and conductivity) using a Meter Group Weather and Soil Condition Station (comprised of a ZL6 Zentra logger system, ATMOS 41 weather sensor system and TERROS12 soil moisture and temperature sensors). Together the deployed combination of surficial efflux monitoring, soil gas sampling, sediment microbiome characterization in conjunction with meteorological parameter monitoring will allow the presence and nature of any CH_4 oxidation within the soils around the examined wellheads to be ascertained.

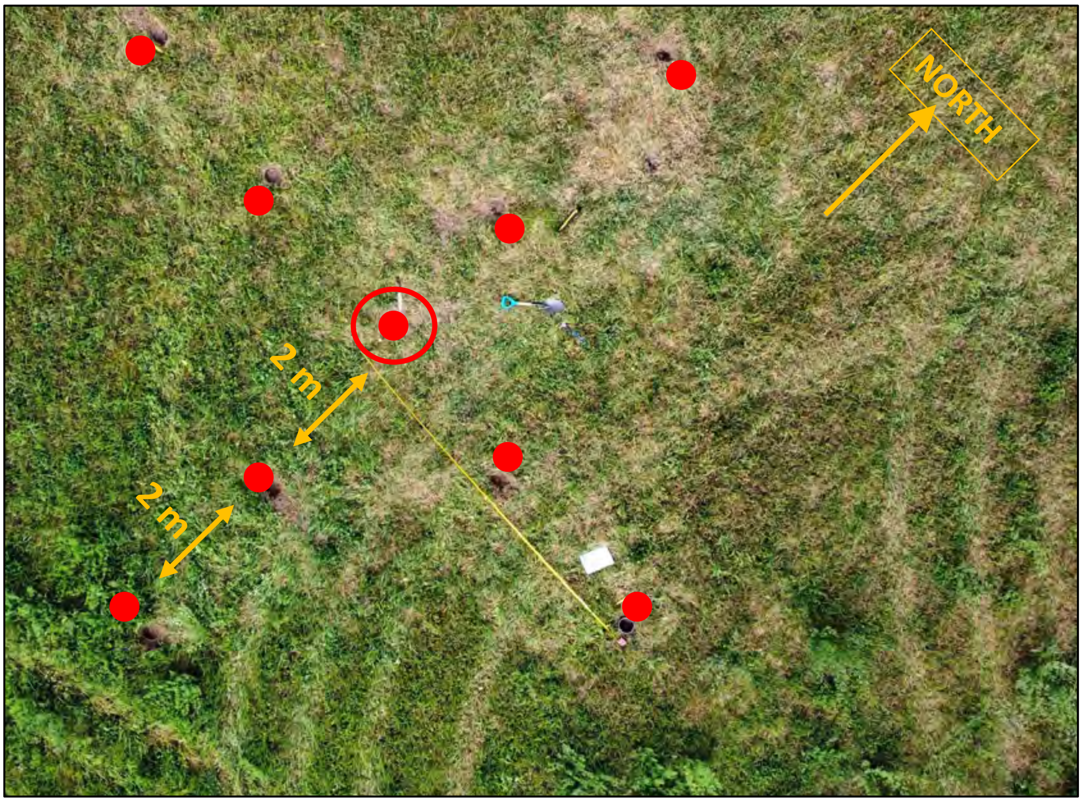


Figure 3: Generalized monitoring grid deployed at each wellsite showing the 9 monitoring points in a cross hair configuration, oriented North, covering an area of 8 m^2 (2 m distance between each point) and centered over the assumed wellhead.

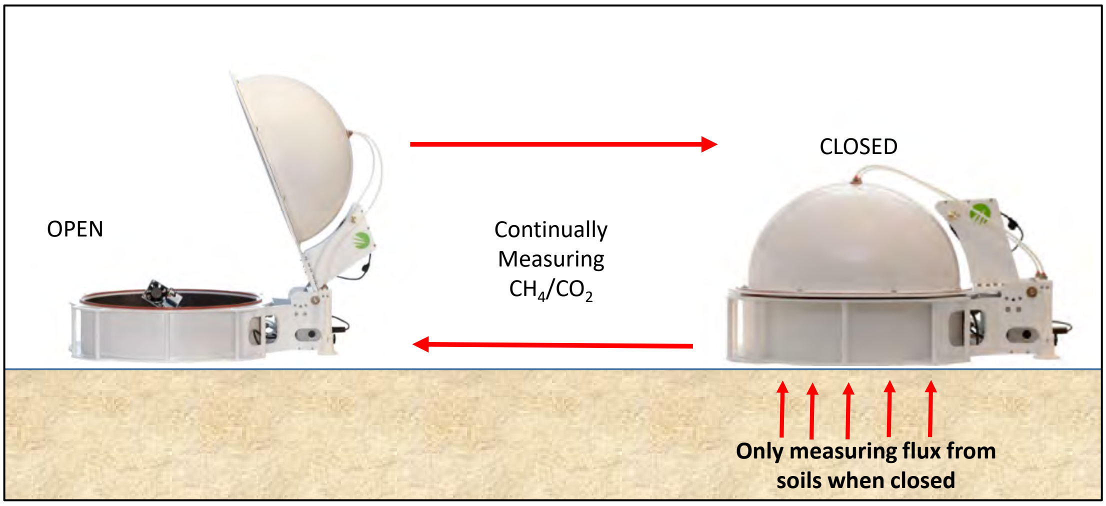


Figure 4: Dynamic flux chamber operational concept showing chamber continually measuring GHG concentrations while opening and closing to estimate fluxes from soils to atmosphere.

Figure 5: Dynamic (left) and static (right) flux chamber systems deployed during field investigations undertaken in 2023 at wellsite WA22800.

3. Results

3.1. Surficial Soils Greenhouse Gas Concentrations and Fluxes

A total of 414 flux measurements were taken across the investigated well sites during the investigation period, including background measurements and measurements at and directly adjacent to the well head (i.e. as part of the monitoring grid outlines in section 2.1). Time series of raw measured CH_4 and CO_2 concentration data (i.e. in ppm) during dynamic flux chamber operation are shown in Figure 6. Time series of associated calculated fluxes are shown in Figure 7 and 8. A summary of calculated fluxes at each wellsite based on observed concentrations is provided in Table 2.

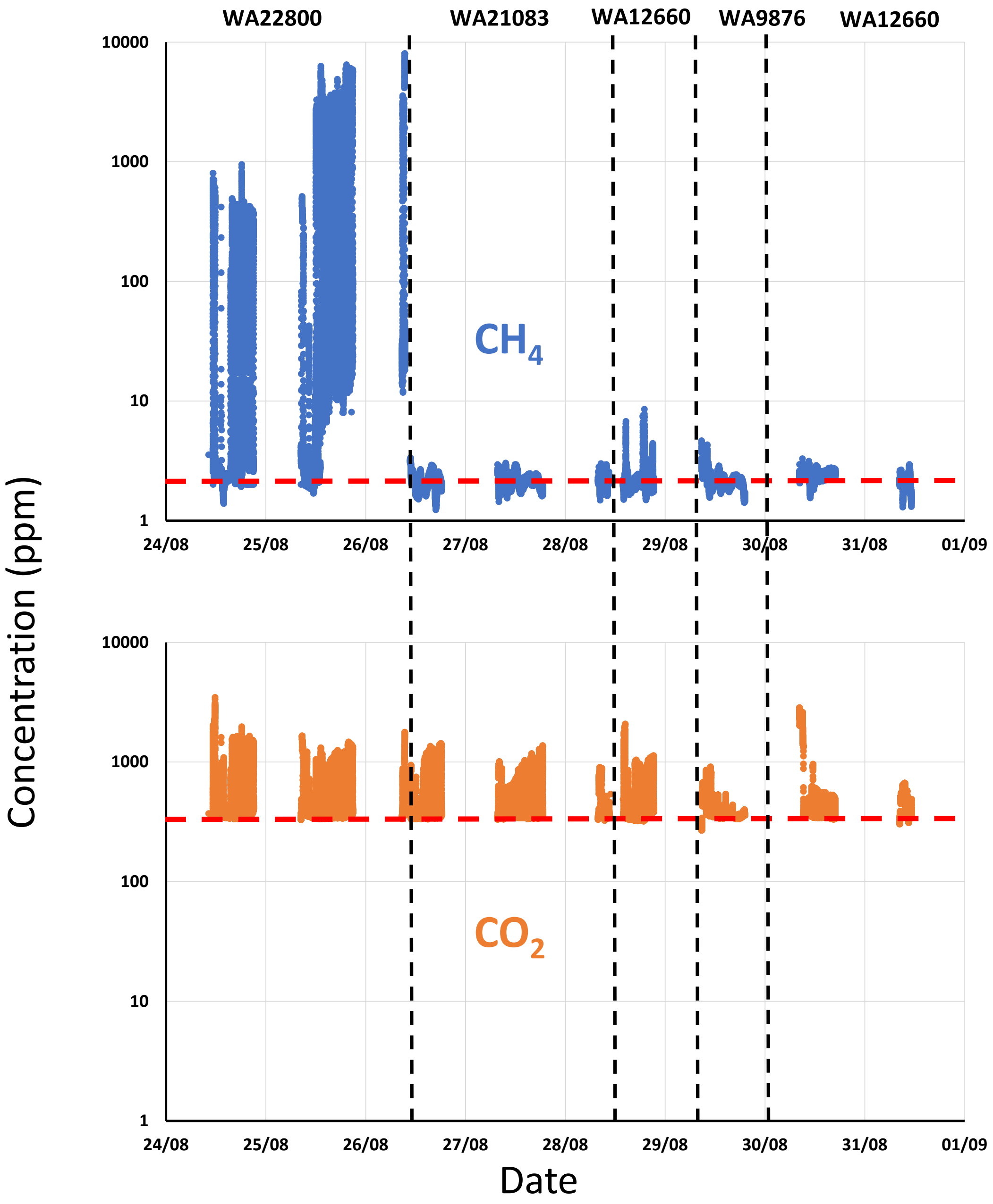


Figure 7: Concentrations of CH_4 and CO_2 as measured with the flux chamber system during field investigation campaign (i.e. throughout all site visits for both open and closed chamber status). The red dashed line shows the typical atmospheric background values for both species. Results show CH_4 was typically low in concentration at all sites with the exception of WA 22800. Meanwhile anomalously elevated CO_2 was clearly detected at WA22800 and again potentially indicated at several other sites where levels frequently exceeded 1500 ppm.

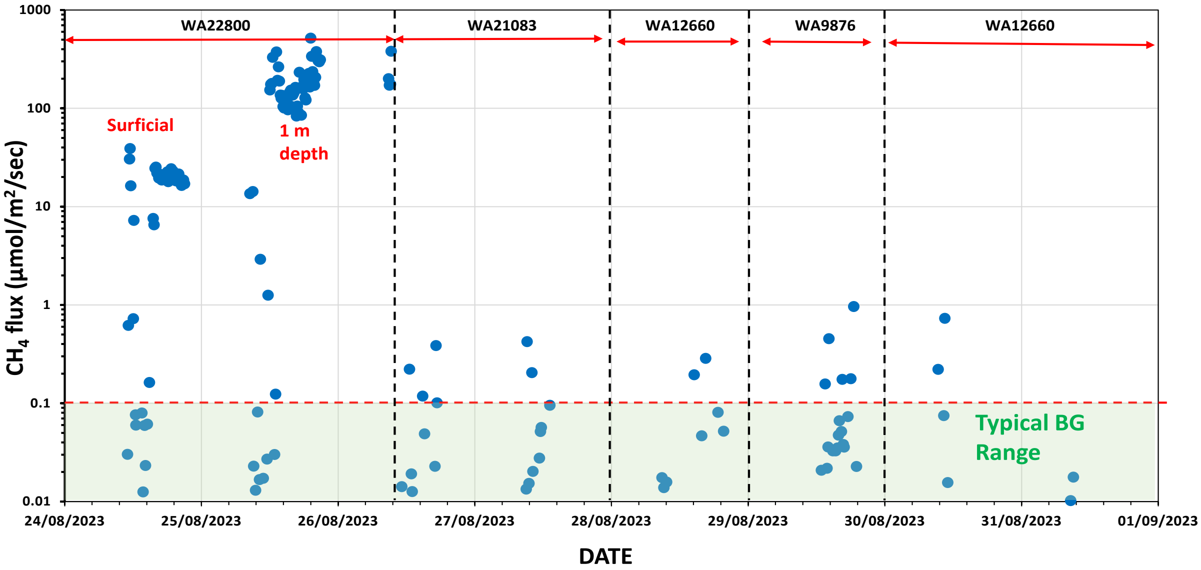


Figure 7: Calculated flux values for CH_4 during the field investigations at all sites (as indicated) with the typical background range for natural CH_4 fluxes indicated by the red dashed line and shaded green area. Results show WA22800 most clearly suffering integrity failure in congruence with the preceding investigations and significantly larger emissions at 1 m depth compared to surface. Meanwhile other sites exhibited only fleeting anomalously high CH_4 fluxes, more typically being within natural ranges.

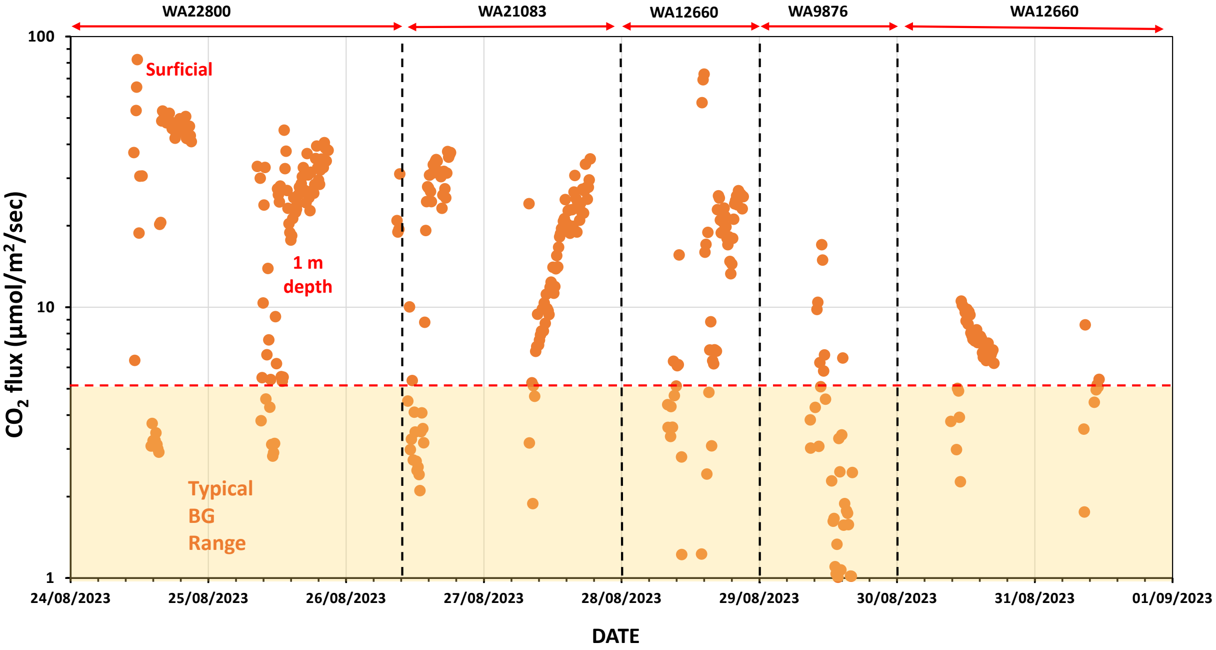


Figure 8: Calculated flux values for CO_2 during field investigations at all sites (as indicated) with the typical background range for natural fluxes indicated by the red dashed line and shaded orange area. Results show WA22800 emitting the most CO_2 in congruence with the preceding investigations and lower emissions at 1 m depth compared to surface. Meanwhile other sites exhibited potentially anomalously high CO_2 fluxes, being commonly in excess of measured natural ranges.

Table 2: Summary of flux estimates at ground surface for each well site examined showing number of flux measurements made, average and maximum CH_4/CO_2 fluxes observed for the wellhead area and background locations. Note for site WA9876 only 1 m depth measurements were made due to time constraints.

Wellsite	**No. of Flux Measurements (BG)**	**Surficial Well Head Area fluxes (μmol/m^2/sec)**				**Surficial Background fluxes (μmol/m^2/sec)**	
		Ave. CH_4	**Max. CH_4**	**Ave. CO_2**	**Max. CO_2**	**Ave. CH_4**	**Ave. CO_2**
12660	12 (5)	-0.007	0.19	23.2	72.7	0.04	6.6
21083	14 (3)	-0.003	0.01	3.7	10.04	-0.02	3.6
22800	61 (9)	12.8	39.0	33.9	82.3	-0.004	3.2

Table 3: Summary of flux estimates at 1 m depth for each well site examined showing number of flux measurements made, average and maximum CH_4/CO_2 fluxes observed for the wellhead area and background locations.

Wellsite	**No. of Flux Measurements**	**1 m Depth Well Head Area fluxes (μmol/m^2/sec)**				**1m Depth Background fluxes (μmol/m^2/sec)**	
		Ave. CH_4	**Max. CH_4**	**Ave. CO_2**	**Max. CO_2**	**Ave. CH_4**	**Ave. CO_2**
9876	35(5)	0.004	0.96	1.3	6.5	-0.04	1.5
12660	28 (3)	-0.03	0.08	21.6	26.2	-0.05	5.7
21083	100 (2)	-0.03	0.42	18.9	37.6	-0.1	2.1
22800	55 (3)	192.3	518.3	28.6	45.1	0.03	5.3

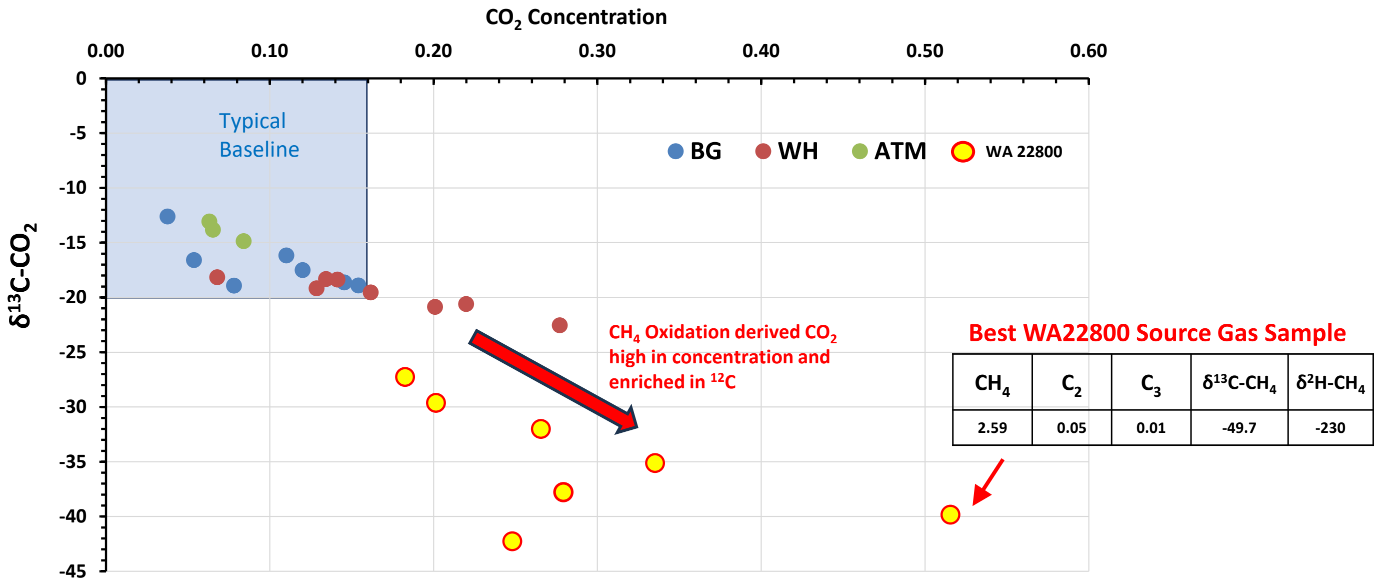
In congruence with the previous year's investigations, well site WA 22800 exhibited the clearest signs of integrity failure with fugitive CH_4 release and concurrent soils-based CH_4 oxidation inferred through excess CO_2 emissions. At WA22800, CH_4 concentrations were seen to be far in excess of what can be considered normal in terms of soil gas CH_4 concentrations exhibiting values as high as ~1000 ppm at surface and ~7000 ppm at 1 m depth. Concurrently, associated CH_4 fluxes to atmosphere at surface of up to 39 μmol/m^2/sec at and around the wellhead were observed, increasing to 518 μmol/m^2/sec at 1 m depth; up to two orders of magnitude greater than observations made at the site the previous year and again clearly indicative that fugitive gas leakage is occurring. Also in congruence with the previous year's results, the three other well sites revisited here exhibited generally more normal soil gas CH_4

concentrations (i.e. ~2 – 6 ppm) and fluxes of CH_4 (i.e. typically < 0. 1 µmol/m^2/sec) at the soil-atmosphere interface. However, it should be noted that during these new investigations that at discrete times and measurement points slightly elevated levels of CH_4 were observed, at times appearing to exceed what might be considered normal (with all three exhibiting several readings in excess of 0.5 µmol/m^2/sec) but not consistently or conclusively. Such observations are marginal, inconsistent and do not indicate conclusive leakage is occurring at these sites, particularly when compared to measurements taken at WA2280 where leakage is incontestable (i.e. CH_4 fluxes > 30 µmol/m^2/sec for most measurements). Thus, based on CH_4 measurements alone, wellsite's WA9876, WA12660 and WA 21083 likely appear to be maintaining full integrity.

Meanwhile, concentrations and fluxes of CO_2 were much more variable across the examined wellsites and seemingly for the most part anomalously high. Here, well site WA22800 where massively elevated CH_4 conditions were identified, was again shown to be co-emitting CO_2 at anomalously high levels (i.e. maximum values of ~80 and ~45 µmol/m^2/sec at surface and 1 m depth respectively compared to background values of 3 – 5 µmol/m^2/sec), almost certainly a result of soil based fugitive CH_4 biodegradation occurring. In addition, of the other three examined well sites, two were seen to be exhibiting what could be considered potentially high CO_2 concentrations and fluxes (i.e. frequently in excess of 20 µmol/m^2/sec, compared to typical background values observed of < 5 µmol/m^2/sec); namely WA12660 and WA21083. In the case of WA12660, CO_2 flux was seen to be as high as ~72 µmol/m^2/sec compared to a background of 6.6 µmol/m^2/sec at surface, whereas for WA21083 CO_2 fluxes of ~37 µmol/m^2/sec compared to a background of 2.1 µmol/m^2/sec were observed at 1 m depth. Interestingly, wellsite WA9876 showed no conclusive signs of elevated CO_2 concentrations or fluxes with an average flux around the wellhead of 1.3 µmol/m^2/sec compared to background measurements of 1.5 µmol/m^2/sec at 1 m depth.

3.2.Soil Gas Laboratory Analyses

Results from physical soil gas sample analyses (i.e. concentrations in ppm and $\delta^{13}C$ values corrected to the international VPDB scale, and stated in units of ‰) taken from investigated well sites and sent for laboratory analyses are shown in Figure 9 for $\delta^{13}C$-CO_2. Atmospheric air samples (taken as controls) are shaded in green, background samples are blue, wellhead samples from W9876, WA12660 and WA21083 are shaded red while samples from WA 22800 are shaded yellow.



CH$_4$	C$_2$	C$_3$	δ^{13}C-CH$_4$	δ^{2}H-CH$_4$
2.59	0.05	0.01	-49.7	-230

Figure 9: Stable carbon isotope ratios for CO$_2$ (corrected to the international VPDB scale, and stated in units of ‰) plotted against CO$_2$ concentrations (in % volume) for samples taken across all sites with atmospheric air samples (green), background samples (blue), wellhead samples from W9876, WA12660 and WA21083 (red) and samples from WA 22800 (yellow). The highest levels of CH$_4$ and CO$_2$ observed in a sample from WA22800 form the best source gas candidate with detailed analytical results for other key parameters associated with this sample shown in the inset table.

Results from laboratory analyses confirm observed field concentrations and flux estimates, showing WA 22800 as exhibiting the highest levels of CH$_4$. Only several samples from WA22800 had concentrations of CH$_4$ high enough to perform a reliable δ^{13}C-CH$_4$ analysis, with one sample in particular (results from which indicated in the inset table in Figure 9) taken from 1 m depth in the soils offering the most promising insight on a potential source of the fugitive gas. For this sample, a δ^{13}C-CH$_4$ of -49.7 ‰ from a concentration of 2.5% volume was observed. This value is in a range typical of deeper, thermogenic gases (which tend to exhibit values of between -50 and -40 ‰), thus inferring a potential thermogenic gas is the source of leakage at site WA22800. In general, as CO$_2$ was detected at much higher levels throughout the investigations at all sites and for all samples, δ^{13}C-CO$_2$ can form a more useful indicator to evaluate for signs and extents of CH$_4$ oxidation at the sites studied. That is, observed δ^{13}C-CO$_2$ can indicate whether the observed anomalously high CO$_2$ is derived from fugitive CH$_4$ oxidation in soils by considering the distinct isotopic signature of the likely carbon source. When fugitive methane is oxidized, the resulting CO$_2$ retains the isotopic signature of the source gas, typically becoming depleted in ^{13}C as microbes performing the process prefer the lighter ^{12}C isotope and preferentially utilize that. Therefore any CO$_2$ generated in this manner (after mixing with the naturally present soil pore space CO_2 derived from organic matter degradation and root zone respiration processes, a $\delta^{13}C$ signature for which is indicated by background samples) leads to development of an increasing-decreasing $[CO_2]:\delta^{13}C\text{-}CO_2$ trend (i.e. an inverse correlation between concentration going up and stable carbon isotope ratio going down). In such cases, $\delta^{13}C\text{-}CO_2$ will converge towards the typically more negative $\delta^{13}C\text{-}CH_4$ signature of the source gas (e.g. 40 - 50‰). In our case, the best source gas sample showed a $\delta^{13}C\text{-}CH_4$ of -49 ‰ while increasing CO_2 levels emitting at WA22800 exhibited increasingly more negative $\delta^{13}C\text{-}CO_2$ values, converging towards the potential source gas and away from the background and atmospheric sample values for $\delta^{13}C\text{-}CO_2$. Here, WA22800 $\delta^{13}C\text{-}CO_2$ signatures were observed to be < -40 ‰ at 0.5 % CO_2 volume compared to background levels of >-20 ‰ at ~0.15% CO_2 by volume and lower as seen in Figure 9. Together, these data strongly indicate that the observed excess CO_2 at WA22800 is from oxidation of a fugitive CH_4 source gas likely of thermogenic nature and represented by the sample taken from 1 m depth (inset table in Figure 9). Data from the other well sites examined are not as conclusive with the observed, more marginally elevated levels of CO_2 around the well heads being much less depleted in $\delta^{13}C$ and only marginally more negative in terms of $\delta^{13}C\text{-}CO_2$ signature in comparison to baseline $\delta^{13}C\text{-}CO_2$ values (e.g. -24 ‰).

3.3.Microbial Analyses

Analyses of 16S rRNA in soils can be instrumental in identifying the presence of methane oxidation around a leaking oil and gas well by characterizing the microbial communities involved in this process. By sequencing the 16S rRNA genes from soil samples, we can theoretically identify and quantify the bacteria present, including methanotrophs, which are microbes that consume methane. In soils near a leaking well, there would likely be an increased abundance of methanotrophs compared to a background site with no methane leakage. This difference in microbial composition and abundance, highlighted by the 16S rRNA analysis, can indicate active methane oxidation due to the elevated methane levels near the well leakage. By comparing the microbial communities of the leaking site to those of the background site, we can assess the extent and impact of methane leakage and the biological response to it. Consequently, we analysed 20 soil samples attained from across the wells sites by sequencing a specific region of their DNA (the 16Sv4 region) using a MiSeq machine. The DNA sequences (Fastq files) generated by the MiSeq were cleaned up for quality and grouped into clusters called operational taxonomic units (OTUs) using the mothur software. Each OTU represents a group of sequences that are at least 97% similar to each other. We detected 3929 different OTUs in the soil samples attained, excluding those that appeared less than 3 times in total. On average, each soil sample had 12,878 high-quality sequences after filtering. We assessed the sequencing quality of both forward (R1) and reverse (R2) reads using the FastQC tool, the results of which are presented in a box-and-whisker plot in Figure 10, which shows the number of high-quality sequences per sample. Only high-quality sequences that matched the 16Sv4 region and were grouped into OTUs were used for further analysis, while any ambiguous or low-quality data were discarded. Each read represents a single DNA sequence fragment obtained from the soil sample. In the context of this study, these reads are specific sequences from the 16Sv4 region of bacterial DNA that are present in the soils at the well sites. Each read provides information about the genetic material of the bacteria present in the soil. By analysing these reads, we can identify and classify the different types of bacteria in the soil samples, based on their DNA sequences and compare them by soil groups. In our case, we differentiate soil samples into three groups by where they were taken including a) "leaking" well group (i.e. soils from around the well head of WA22800), b) "potentially leaking" wells group (soils from around the well heads of other wells examined), and, c) "not leaking" (background samples from all sites). These groupings can now be used to evaluate for differences between soil microbiomes.

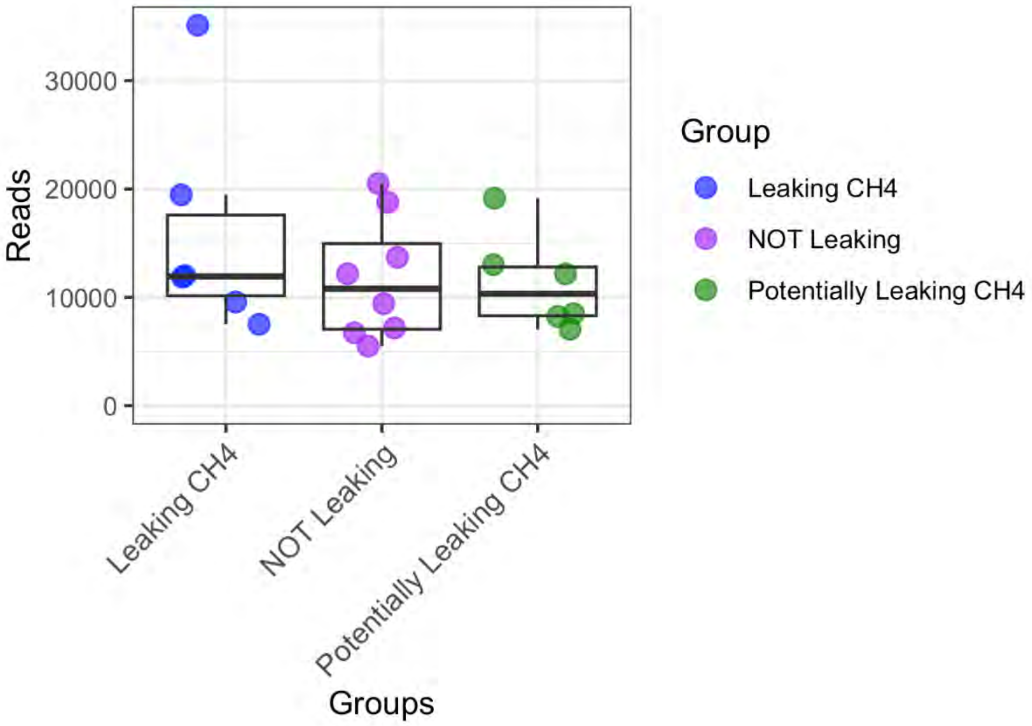


Figure 10: Box and whisker plot of reads per soil sample by group showing similar levels between groups, with a propensity for higher reads in several leaking well soils (i.e. collected around WA 22800).

The number of reads typically obtained from natural soil samples can vary widely depending on several factors, such as the complexity of the soil microbial community, the sequencing depth, and the methodology used. However, a common range for high-throughput sequencing of soil samples is between 10,000 to 100,000 reads per sample. In our specific study, we attained an average of 12,878 quality-filtered reads per sample, which falls within the typical range for soil microbiome studies. This number is sufficient to capture a representative snapshot of the microbial diversity present in the examined soils and evaluate for CH_4 oxidation processes.

Next, high quality reads were classified using Silva v. 138.1 as the reference database. We aggregated OTUs into each taxonomic rank, and plotted the relative abundance of the most abundant ones. Results showed that the relative abundances of the most abundant phyla and genera appear to be quite different across groups. The aggregated taxa were visualized at the family level taxonomic rank and are shown in the following taxonomic bar plots in Figure 11. The unfilled portion of the bar plots represent lower-abundance taxa.

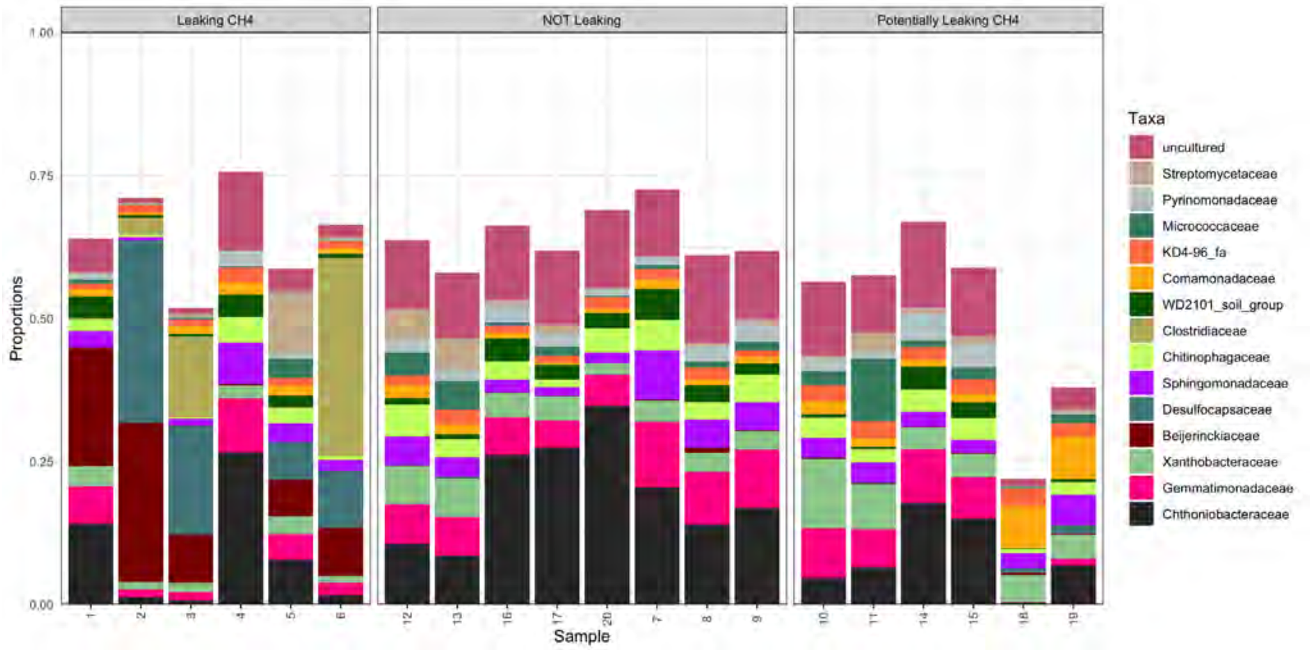


Figure 11: Aggregated taxa from soil samples visualized at the family level taxonomic rank with the unfilled portion representing lower-abundance taxa. Data show differences between the groups of soils with several families including Beijerinckiaceae and Desulfocapsaceae being more prominent within the leaking well soils.

Next, the Shannon Index was calculated which is a measure of biodiversity that accounts for both the abundance and evenness of species in a soil community. The Shannon Index is calculated based on the proportion of each species present in a sample and helps compare microbial diversity across different samples or conditions, monitor changes over time, and gain insights into the health and functionality of soil ecosystems. A higher Shannon index suggests a more diverse and potentially more resilient microbial community, which is crucial for maintaining soil health and performing various ecological functions. This metric provides valuable information about the impact of environmental changes on microbial communities, helping understand and manage soil ecosystems more effectively. In our case, we are looking to see if there are differences between the soil groups, with focus on differences between soils at the leaking wellhead (i.e. those that are being exposed to high levels of methane) and soils at background locations away from leakage. Results show a generally lower, statistically significant difference in the Shannon Index for soils at the leaking well (average SI = 4.1 and as low as 3) compared to the background locations (average SI = 5) and potentially leaking wells (average SI = 5). This is likely because the introduction of methane at the leaking well (i.e. WA22800) has led to proliferation and domination of specific methane-oxidizing bacteria, reducing the overall evenness and richness of the microbial community. The dominance of CH_4 oxidation associated microbes likely decreases the diversity of the community, resulting in a lower Shannon Index at the leaking well. In contrast, the background samples and seemingly the potentially leaking wells, are unaffected by methane leakage, and appear to exhibit a more diverse and evenly distributed microbial community, reflected in a higher Shannon Index.

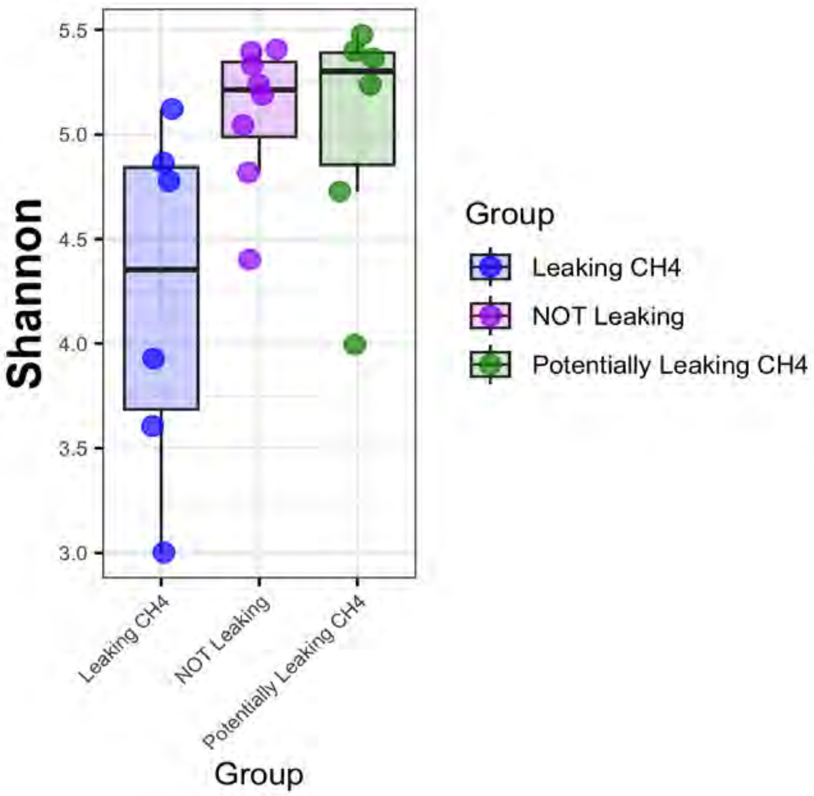


Figure 12: Box and whisker plot of Shannon Indices for analyzed soil samples by group. Results show a statistically significant difference between the leaking and not leaking soil groups, highlighting the occurrence of CH_4 oxidation in the leaking wellhead soils and indicating that it profoundly alters the soil microbiome.

Finally, the *Linda* function from MIcrobiomeStat package was used to identify differentially abundant taxa among Group variables. This approach involves statistically analyzing the microbial communities to detect which specific bacteria are significantly more or less abundant in each group. By comparing the microbial composition of soils affected by methane leakage to those from background sites, we can pinpoint the taxa that are particularly responsive to the presence of CH_4 and likely involved in its oxidation. This analysis reveals how microbial communities change in response to CH_4, highlighting the specific bacteria that may play a role in methane oxidation or other related processes. A linear model on centred-log ratio transformed data (i.e. log fold 2) was used for the test to evaluate the leaking and not leaking soil groups to see what was most different about them in terms of microbiome with the outcome shown in Figure 13. In addition, the difference in key taxa derived from the 16S rRNA analyses at the family level are summarized in Table 4. Results show that a range of microbial groups are present at much higher levels in the leaking well soils group compared to the background soils group samples. Here of note are the genera Beijerinckiaceae and Methylomonadaceae, both of which have been linked to oxidation of CH_4 in environmental settings[17-19]. Here their larger presence within the leaking well site soils is indicative that they are involved in the observed CH_4 oxidation processes and we suggest they should be more carefully considered to understand their role and the mechanisms employed.

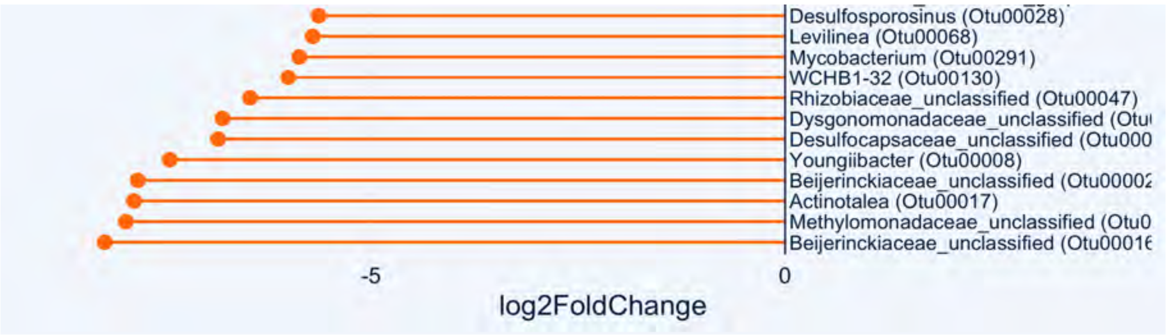


Figure 13: Difference in abundance of key taxa between “leaking” and “not leaking” group soil samples at the genus level showing some groups are far in excess within the soils around the leaking well. Some of these are linked to CH_4 oxidation in natural settings and should be more carefully considered to understand their role in natural soils CH_4 oxidation and the mechanisms employed.

Table 4: Microbiome % as measured by 16S rRNA analyses formed by key taxa at the family level for samples from the wellhead and background locations at WA22800.

Sample Location	Methylomonadaceae	Beijerinckiaceae	Desulfocapsaceae	Cumulative (%)
WA22800 Well Head	**1.81**	**11.93**	**11.24**	**24.97**
WA22800 Background	**0.03**	**0.35**	**0.04**	**0.42**

3.4.Meteorological Conditions

Time series of key weather parameters (i.e. air temperature, barometric pressure and wind speed) that prevailed during the field campaign as monitored by the deployed weather and soil sensor system during the investigations are presented in Figure 14. Days on which individual wellsites were investigated and the prevailing conditions are indicated with dotted red lines and numeric labels.

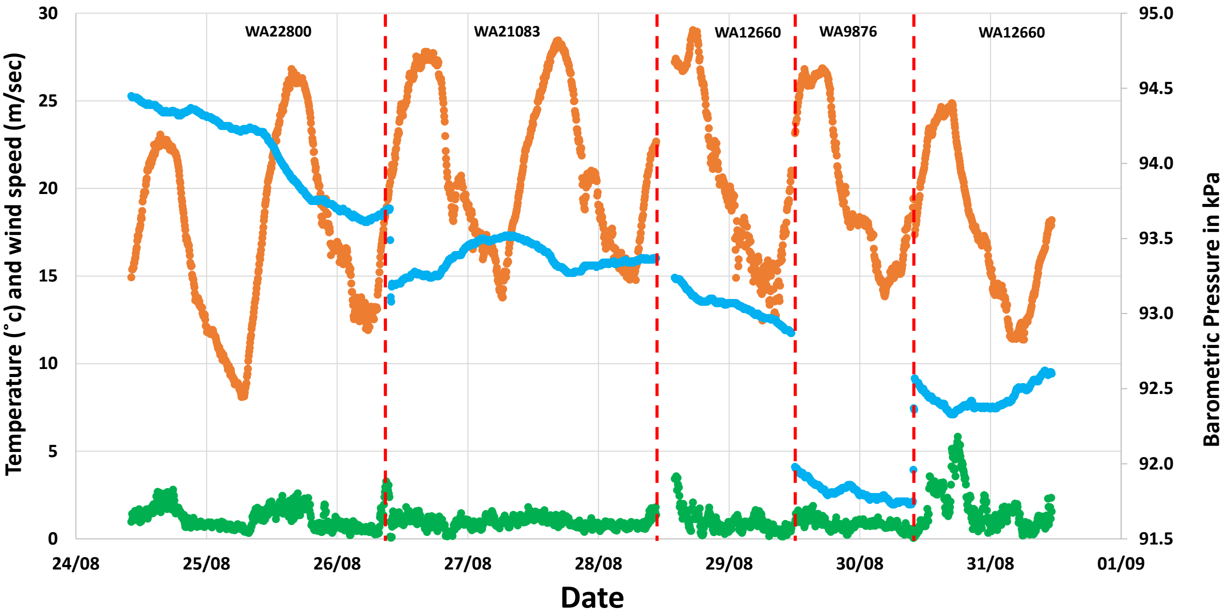


Figure 14: Key weather parameters wind speed (green series), air temperature (orange series) and barometric pressure (blue series) during field investigations with days each well site was visited indicated in by labels and delineated by red dashed lines.

Results show prevailing weather conditions were typical of Northeast BC in late August with generally warm (including normal diurnal temperature maxima and minima) and dry conditions (i.e. no rainfall occurred several days before and during the survey period) with minimally fluctuating wind speeds (generally quite low in magnitude, typically less than 2 – 3 m/sec). These data show that conditions were only marginally variable by day of the field investigation, which is unlikely to have had a significant impact on observed results. Of note is that barometric pressure was observed to continually decline as field investigations proceeded, albeit at a gradual rate, bottoming out on the last days of the campaign, a behavior which could have affected some observations, however in which manner is not immediately clear and not expected to change any findings or conclusions made.

3.5.Field Based CH_4 Oxidation Rate Estimations

Of key interest in the current project is to better quantify and constrain potential natural soils-based CH_4 oxidation rates with newly collected field data. From the new observations made in this year's field campaign (as described previously), wellsite WA22800 showed huge potential to generate useable and robust soils based CH_4 oxidation estimates. This was particularly the case due to the flux data collected both at ground surface and 1 m depth at the same point above the wellhead. These data showed very clearly a significant level of CH_4 oxidation occurring based on the reciprocal, inversely correlated fluxes of CH_4 and CO_2 with depth through the soil column and as supported by stable carbon isotope and microbial data. Here, surficial (i.e. at ground surface) flux observations showed higher fluxes of CO_2 (i.e. ~50 µmol/m^2/sec) with lower (but still significantly in excess of natural ranges) fluxes of CH_4 (i.e. ~20 µmol/m^2/sec) with a CO_2:CH_4 ratio of ~2.5. However, at 1 m depth, this trend was reversed with CH_4 now being by far the major gas flux (i.e. ~100 - 300 µmol/m^2/sec) with lower levels of CO_2 flux (i.e. ~30 - 40 µmol/m^2/sec) and a CO_2:CH_4 ratio of ~0.17. These data are shown in Figures 15, 16 and 17.

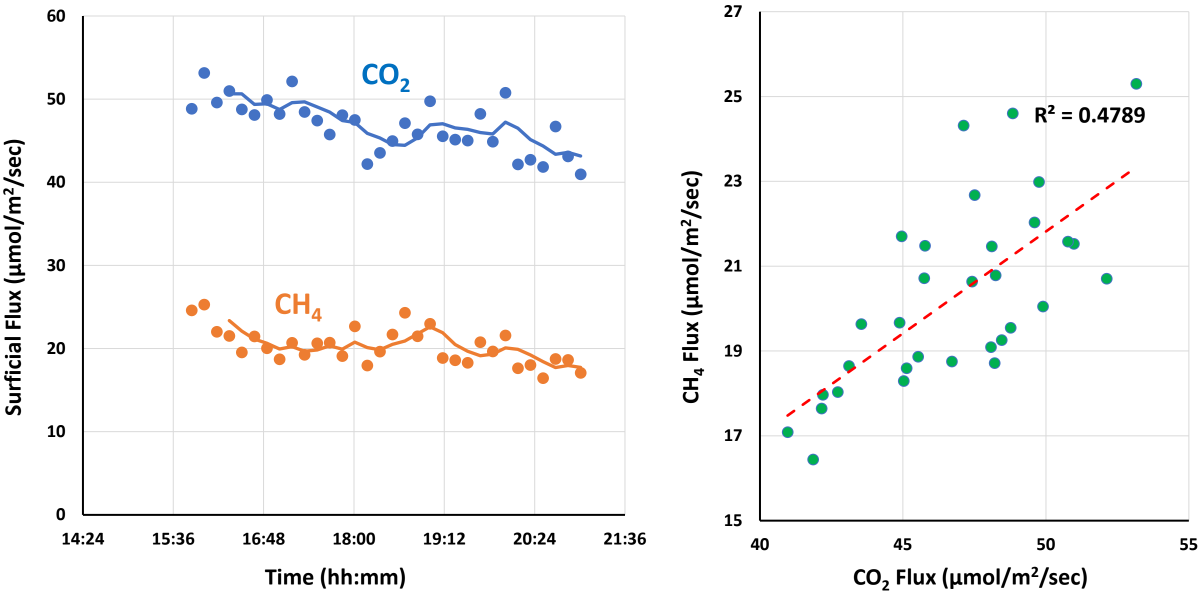


Figure 15: Surficial continuous flux data as observed on 24/08/2023 above the wellhead at site WA22800 showing CO_2 as the dominant emitting gas with CH_4 approximately half the magnitude, with both gases reasonably correlated, highly indicative of the CO_2 being derived from CH_4 oxidation.

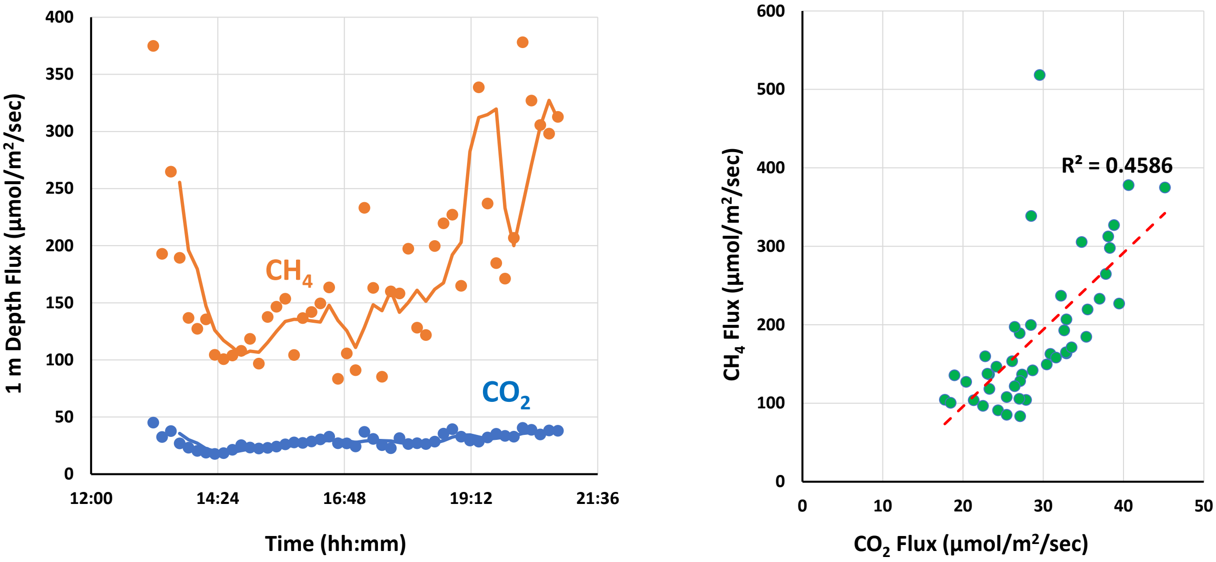


Figure 16: Observed fluxes at 1 m depth as observed on 25/08/2023 above the wellhead at site WA22800 (i.e. beneath the exact location where surficial fluxes were measured as shown in Figure 15) showing that beneath 1 m of soil column, CH_4 forms the dominant emitting gas. Here, CO_2 is approximately 20% the magnitude observed at the surface, with both gases again reasonably correlated, highly indicative of fugitive CH_4 being oxidized through the soil column.

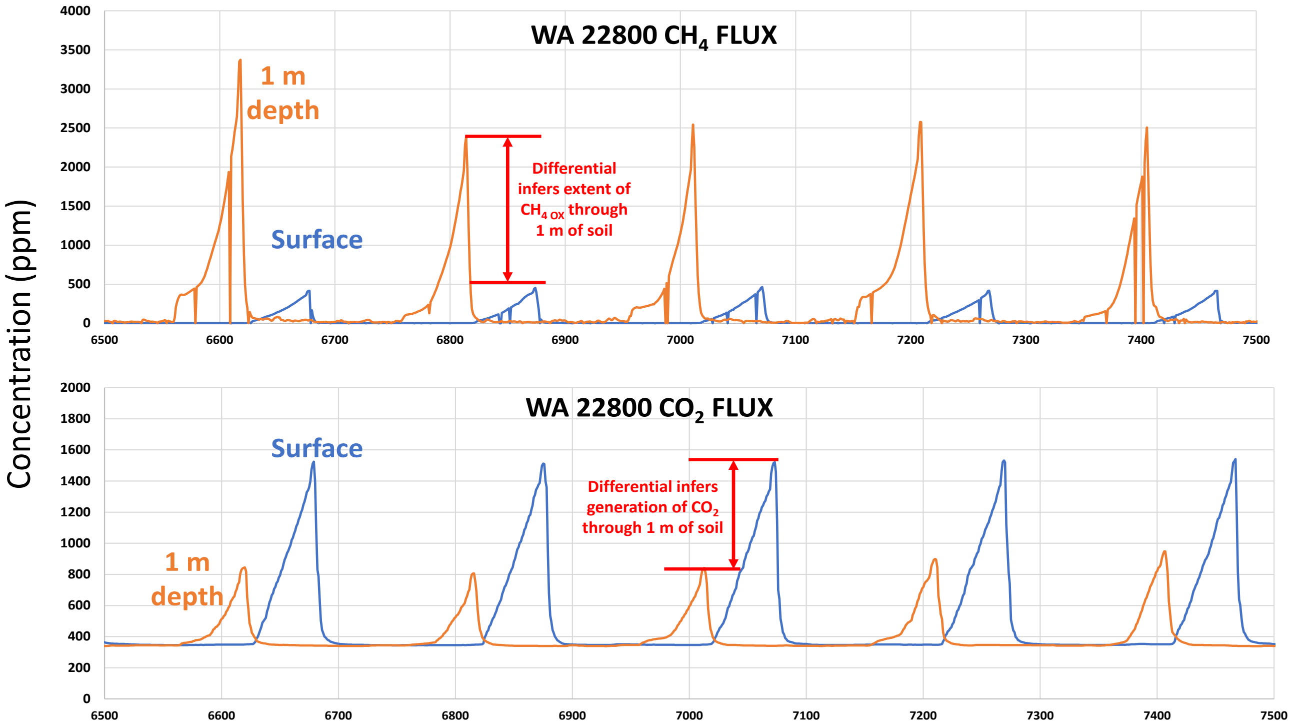


Figure 17: Generic time series of CH_4 and CO_2 concentrations during flux measurements at surface and 1 m depth showing the reciprocal relationship between the species whereby CH_4 maximal concentrations decrease at the surface while CO_2 increases inferring the loss of CH_4 is associated with soils-based oxidation from which an oxidation rate estimate can be made.

Such data are valuable as they can be used directly to infer a field based CH_4 oxidation estimate by considering the reduction in CH_4 over 1 m vertical travel distance of fugitive CH_4 through the soils. That is, by considering the flux of CH_4 at 1 m depth compared to surficial flux observations we can infer how much mass has been removed per unit time and volume/area of soils to generate an in-situ observed CH_4 oxidation rate. Thus, field measurements of CH_4 and CO_2 fluxes at surface and 1 m depth were used to better constrain the CH_4 oxidation rates occurring. Here, field data collected over two days, including data from 24/08/2023 (consisting of 32 measurements) taken from the soil surface and from 25/08/2023 (consisting of 47 measurements) taken from 1 m depth were combined. For the purposes of estimating oxidation rates from this dataset, it was necessary to introduce the assumption that these two sets of measurements were acquired simultaneously, i.e. the surface reading from day 1 was measured at the same time as the 1 m depth reading from day 2. By introducing this assumption, we were able to pair a methane flux at surface with a methane flux at 1 m depth and determine a mass removed per time. Following a conversion to account for the mass of soil in which the oxidation rate is occurring, the oxidation rate was then derived as the difference between these values (i.e. the difference between what goes into the soils at 1 m depth and what comes out at surface).

Methane oxidation rates estimated from this approach are shown in Figure 18 and can be seen to vary from 60 to 530 µg /g_{soil}/day with an average of 195 µg/g_{soil}/day (which equates to an average of ~230 g CH_4 /m^2/day assuming a constrained reactive soil area of $1m^2$ and soil bulk density of $1300kg/m^3$).

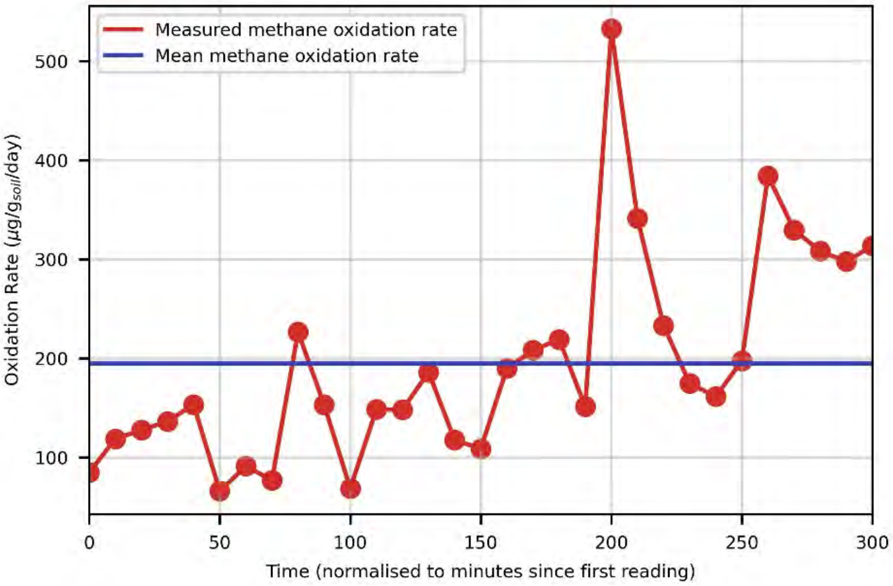


Figure 18: Plot of oxidation rates derived from field measurements with the average value (195 µg/g soil/day) also plotted.

4. Discussion and Conclusions

Here we described a new, bespoke field investigation conducted at key wellsites of interest to better understand CH_4 oxidation in natural soils of BC, with a view to estimating oxidation extents/rates and identifying microbial communities that may be performing the oxidation process. The following are the main findings and observations from these field investigations and newly collected data:

- Observations were consistent with the previously conducted fieldwork, showing clearly anomalous CH_4 and CO_2 emissions at wellsite WA22800, with only seemingly anomalously elevated CO_2 emissions (i.e. with no excess or anomalous CH_4 present) at the other well sites examined. This observation suggests consistency inter-annually for wells suffering leakage.
- The integrity status of WA9876, WA12660 and WA21083 remains uncertain. While previously anomalous levels of CO_2 were detected at each site, and these new investigations further identified seemingly anomalous levels of CO_2, no excess CH_4 has been further detected (despite advancing measurements to 1m depth into soils at and around the well head) and stable carbon isotopes for detected CO_2 appear inconsistent with derivation from thermogenic CH_4 oxidation. More work is needed to constrain the integrity of these wells. Consequently, most

focus was placed on WA22800 in the subsequent data analyses and interpretations, as data from this site was most useful to characterize CH_4 oxidation processes, reflected in the following highlights.

- Aside from general consistency in observed CH_4 and CO_2 emission profiles year on year, wellsite WA22800 exhibited 10-fold higher CH_4 fluxes from ground surface into the atmosphere this year compared to last, with average and maximum surficial CH_4 fluxes around the wellhead of ~13 and ~39 µmol/m^2/sec in 2023 compared to 0.7 and 3 µmol/m^2/sec in 2022. Moreover, once the soils at WA22800 were penetrated to 1 m depth during these new 2023 investigations, CH_4 fluxes were seen to be even higher at 200 and 500 µmol/m^2/sec (a 1000 times higher than previously detected at surface) for the average and maximum observations (i.e. several hundred times greater than observed at the surface in 2022). Meanwhile, the co-emission ratio of CO_2:CH_4 inverted with depth whereby much less CO_2 was observed emitted at 1 m depth compared to from the soil surface.
- The average oxidation rate estimate for CH_4 in soils at WA22800 was 195 µg/g_{soil}/day (which equates to an average of ~230 g CH_4 /m^2 of soils/day assuming a constrained reactive soil area of 1m^2 and soil bulk density of 1300kg/m^3). This value is high and exceeds levels reportedly attained in engineered landfill biocovers[20], suggesting that natural soils have a propensity for extensive CH_4 oxidation (at least in this location and under the seasonal conditions prevailing) and warrants further investigation and understanding to confirm and validate.
- Stable carbon isotope results for CH_4 and CO_2 gases detected strongly supports that fugitive CH4 at WA22800 is of a thermogenic nature and that elevated levels of CO_2 are derived from fugitive CH_4 oxidation. However, other stable carbon isotope data collected only weakly support that elevated CO_2 at other sites examined is potentially associated with fugitive CH_4 oxidation.
- Microbial analyses (i.e. 16S rRNA) showed that soils at the leaking wellhead were microbially distinct (in a statistically significant manner) from soils in background locations and also from potentially leaking well soils. Leaking well soils were less diverse, being dominated by several key taxa including Beijerinckiaceae and Methylomonadaceae at the family level that are therefore likely involved in the CH_4 oxidation process and have been linked to CH_4 oxidation in environmental settings in other studies.

WP3: Reactive Transport Modelling

1. Introduction and Background

In this work package, insights gained from WP1 and WP2 were used to perform reactive transport modelling to simulate CH_4 oxidation processes in soils at and around integrity compromised well WA22800. The generalized modelling concept was to integrate newly attained information on CH_4 oxidation rates from the field (i.e. as described in WP2), assuming gas leakage volumes and other controlling factors/parameters such as temperature variation, soil moisture content, soil composition/permeability in addition to any other important BC specific properties to simulate reactive CH_4 transport in soils. Key to recognize is that methane oxidation rates within soils (often studied at landfill sites), are known to be strongly influenced by the soil temperature and moisture content[21], which are in turn determined by the surface climate/seasonal conditions and the properties of the soil. Thus, a coupled climate and transport model is required to account for these aspects and meaningfully simulate CH_4 oxidation in soils. Therefore, we selected a model that is well-established in the field of estimating landfill methane emissions, but to our knowledge, has not been utilised with the purpose of estimating CH_4 leakage from abandoned wells (considered analogous to buried waste within this context), namely CALMIM (the California Landfill Methane Inventory Model). CALMIM is a 1-dimensional transport and oxidation model that evaluates landfill methane emissions, including oxidation processes and accounts for site characteristics (soil and engineered covers) as well as simulated weather conditions. CALMIM was originally developed for the California landfill methane inventory[22-24] and its accuracy has since been tested and validated using various field measurements[25-27]. It is currently deployed as an IPCC (Intergovernmental Panel on Climate Change) Tier III model for methane emissions from solid waste disposal sites. It was developed as a JAVA application, which is freely available to download from the USDA website.

2. Methods

2.1. CALMIM Model Overview

CALMIM ultimately predicts diffusive methane emissions over an annual cycle for a given location, while accounting for methane oxidation. It uses a 1-dimensional finite difference model (adopting Fick’s law) to calculate heat, water and bidirectional diffusive gas transport (i.e. CH_4 and O_2) through defined cover materials (i.e. engineered covers or natural soils of various compositions). The model is designed to address site-specific operational factors such as soil composition and gas concentrations with depth (i.e. through a lower boundary condition), at surface (an upper boundary condition) and site-specific climate factors (such as temporal evolution of soil moisture and temperature). The climate model is linked to an embedded USDA global climate model based on 30-year climate data that inputs the average weather conditions to help predict how methane oxidation in soils will proceed through the seasons based on geographical location. A full detailed overview on how CALMIM is constructed and works can be found the associated United States Department of Agriculture (USDA) website. Overall, general inputs to the CALMIM model can be broadly classified into three categories: (i) site characteristics, (ii) cover material (i.e. soil) characteristics, and, (iii) boundary conditions. The inputs for each of these categories used in the current modelling of CH_4 oxidation at WA22800 are outlined below.

2.2.Model Input Data and Boundary Conditions

2.2.1. Site Characteristics

For the purposes of this modelling study, we focused on site WA22800 located near Cecil Lake and the observed constrained surface leakage footprint for which a generalised site area of $1m^2$ was assumed with all associated input parameters shown in Table 1. The climate model embedded within CALMIM for this site (based on provided coordinates) includes a mean air temperature range from ~ -15 °C in the Winter to 15 °C in the Summer, with precipitation peaking around June at 12 mm per day (as shown in Figure 1). During the winter months, this region can usually experience several feet of snowpack and frozen ground, however to our knowledge, CALMIM does not account for this phenomenon in its transport model.

Table 1: Site Characteristics used as input to the WA22800 CALMIM simulations.

CALMIM Input Parameter (unit)	Value
Latitude (decimal degrees)	56.3047
Longitude (decimal degrees)	-120.5753
Approx. Surface Footprint (acres)	2.5×10^{-4} (equivalent to $1 m^2$)

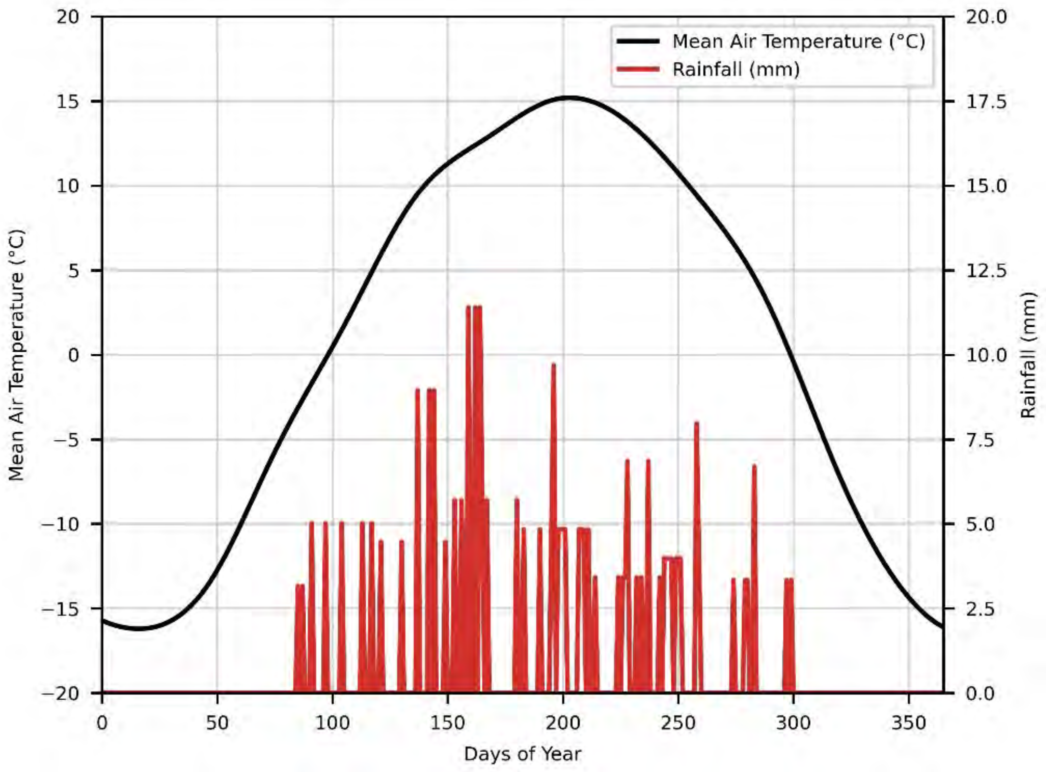


Figure 1: Mean air temperature and rainfall estimated for the area of WA22800 according to the USDA global climate model that is embedded in CALMIM.

2.2.2. Soil Characteristics

The characteristics of WA22800 soils (termed cover in the CALMIM model) were input directly from field measurements and observations made during 2022 and 2023 field investigations. The deepest CH_4 reading obtained from 1 m depth was selected as the lower boundary of the model and soil layer thickness set to match. Soil characteristics employed in the model are summarized in Table 2.

Table 2: Soil characteristics used as input to CALMIM simulations for WA22800 based on field observations.

CALMIM Input Parameter (unit)	Value
Coverage (%)	100
Organic matter (%)	3.3
Vegetation Presence (%)	80
Gas Recovery System	N/A
Cover Layer Material	Silty Loam
Cover Later Thickness (m)	1

2.2.3. Boundary Conditions

Key boundary conditions used in the CALMIM model simulations were also based on field observations alongside best-case assumptions for the region/area and are summarized in Table 3.

Table 3: Boundary Conditions input to the CALMIM simulations.

CALMIM Input Parameter (unit)	Value
Air (upper boundary) temperature (°C)	(generated from climate model)
Waste (lower boundary) temperature (°C)	10
Surface (upper boundary) moisture (v/v)	(generated from climate model – precipitation)
Waste (lower boundary) moisture (v/v)	Assumes gravity drainage
Surface (upper boundary) methane concentration (%)	0.0002
Waste (lower boundary) methane concentration (%)	43
Surface (upper boundary) oxygen concentration (%)	20
Waste (lower boundary) oxygen concentration (%)	18.5
Maximum methane oxidation rate µg/g_{soil}/day	195

2.2.4. Consideration of Variable Oxidation Rates

Key to recognize is that parameter “maximum methane oxidation rate” strongly determines all model outputs and so needs to be carefully considered and constrained. To demonstrate the sensitivity of model results to this parameter, all other values (i.e. as stated above with the exception of oxidation rate itself) were fixed, and multiple simulations were undertaken with varying methane oxidation rates including 1, 5, 10, 15, 20 and 25 µg/g_{soil}/day (Figure 2). Based on these inputs the resulting modelled surface emissions were seen to be hugely variable during the summer months, ranging from ~ 8 g/m^2/day to zero as shown in Figure 2.

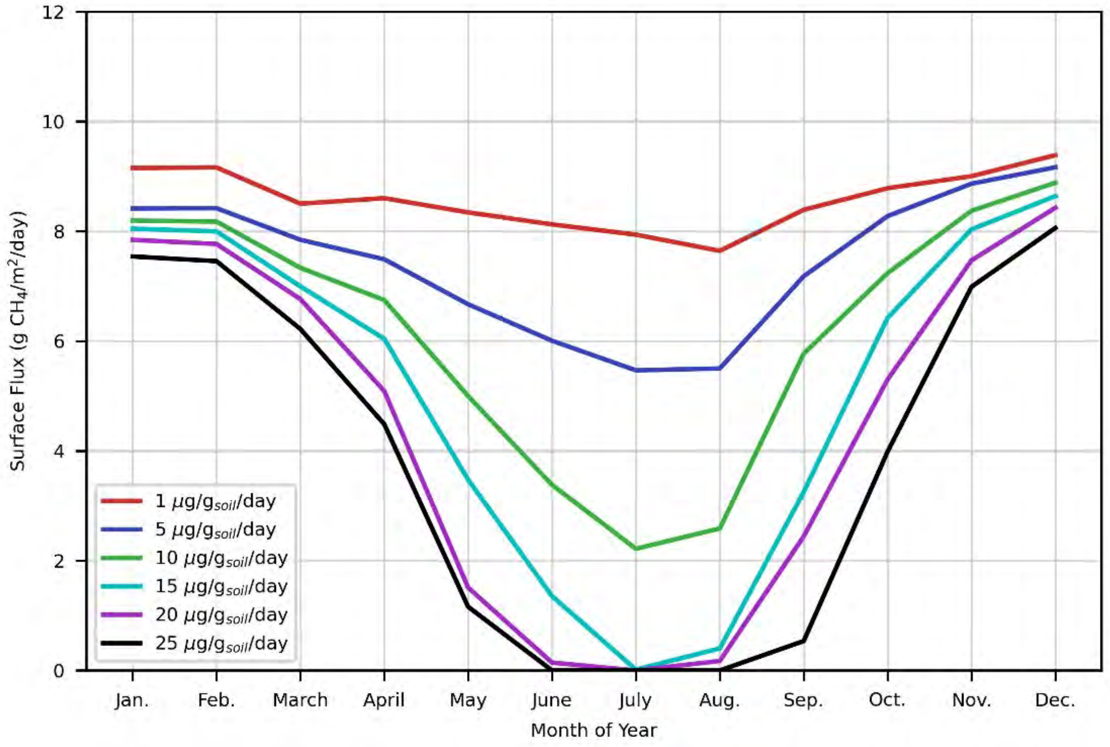


Figure 2: Modelled surface methane fluxes for various maximum methane oxidation rates to demonstrate dominance of key parameter on model outputs.

3. Results

3.1. Modelling Scenario 1: Adjusting CH_4 Lower Boundary Concentration

The CALMIM model works according the principle of diffusive flux, based on the concentration gradient between the upper and lower boundary conditions of CH_4. Thus, it does not include an advective (i.e. pressure driven) flux component which forms a slight limitation of the (primarily landfill focused) model with respect to integrity compromised petroleum well fugitive gas leakage simulation. For leaking oil and gas wells, typically it is expected that advective (i.e. pressure driven) fluxes are the main driver for fugitive gas migration and not diffusion. Consequently, it was expected that in order to replicate the observed surface fluxes at site WA22800 meaningfully it would be necessary to artificially increase the lower boundary in order to get higher fluxes representative of both advective and diffusive fluxes as were being observed at the site. Thus, during this exercise, a series of simulations were conducted to better constrain a suitable 1 m deep methane concentration value (i.e. the lower boundary condition of the model) that could produce the CH_4 surface fluxes observed in the field (i.e. 34.6 g/m^2/day during the month of August) considering all the input values listed in Tables 1, 2 and 3. Here, the monthly average methane emissions for various lower boundary methane concentrations (at 5 % volume increments) were simulated (Figure 4). From analysis of these, it was apparent that to approach the field measured values of flux, a lower boundary of 40 - 45 % CH_4 by volume was necessary (as opposed to values of 2.5% measured in the field), therefore an additional simulation at 43 % was undertaken to provide a close fit. Results from this exercise are shown in Figure 3 where the field observation is shown as a black circle that corresponds best with the 43% lower CH_4 boundary condition. Of note is that all modelled surface fluxes exhibit a strong seasonal trend with oxidation occurring mostly during the summer months and oxidation seemingly not occurring during winter periods as shown by the inversely correlated CH_4 emission and oxidation masses that were simulated (i.e. Figure 3). Here, for all lower boundary concentrations examined, at least 100 g CH_4/m^2/day emissions were simulated as being emitted during the winter months, and in the case of the 43 % lower boundary concentration (i.e. that most relevant to our field observations) peak emissions of ~150 g CH_4/m^2/day were estimated during December when CH_4 oxidation was assumed not to be occurring. All simulations showed a steady decline in emissions during spring, with ~ 30 g CH_4/m^2/day oxidation during April resulting in ~ 110 g CH_4/m^2/day of emissions. This was followed by a much steeper decline during the summer, to only around ~13 g CH_4/m^2/day of emissions during July and a corresponding peak of ~ 130 g CH_4/m^2/day oxidised. As these values are monthly averages, we up-scaled each by the number of days in the month (i.e. for 2023) to obtain the overall total mass of methane emissions in g/m^2/month (Table 4). Total annual emissions were then calculated as the sum of monthly values, estimating a yearly-modelled value of 35.15 kg of CH_4/m^2, compared to a field observation based estimate of 12.6 kg of CH_4/m^2 based only on observations made in August (determined by extrapolating the measured field emissions values for an entire year). Although interesting to highlight, this result is not necessarily surprising or unexpected considering field data corresponds only to the summer months (i.e. late August) when oxidation is expected to be higher, and therefore emissions would be lower. However, here we show with a well-recognized and validated reactive transport model that oxidation rates would be very low to non-existent in winter and maximal during summer conditions. In our case, we only have data from late August (i.e. peak potential oxidation during summer conditions) and so it would be desirable to further validate this phenomenon with respect to fugitive gas leakage by attaining measurements from WA22800 (and other sites like it) in differing seasons including spring, fall and winter.

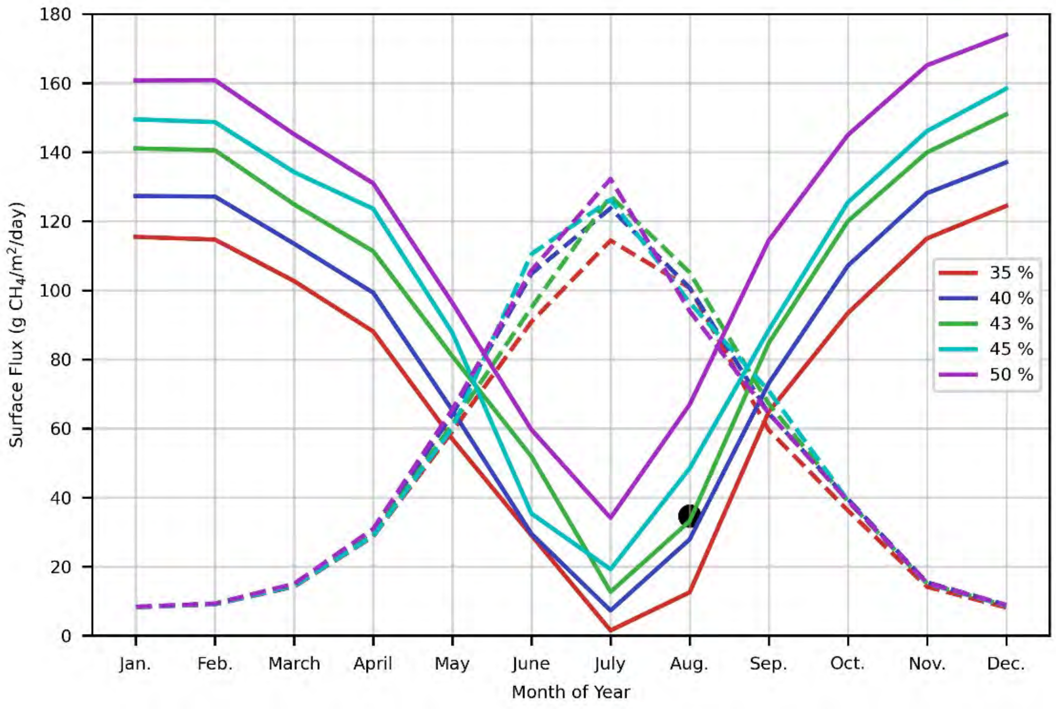


Figure 3: A series of simulation results showing the average, monthly methane surface emissions, with oxidation (solid lines) and average, monthly, oxidised methane (stippled lines) for a range of methane lower boundary concentrations. The black data point represents the field-measured flux value (i.e. from August 2023 of 34.6 g $CH_4/m^2/day$).

Table 4: Modelled monthly CH_4 emissions for the simulation case with a 43 % concentration of CH_4 at the lower boundary (providing the closest fit with the field measured fluxes).

Month	**Emissions (kg/m^2)**
January	4.37
February	3.94
March	3.87
April	3.34
May	2.51
June	1.56
July	0.39
August	1.03
September	2.54
October	3.72
November	4.20
December	4.68
Annual	**36.15**

To better understand which factors are driving the modelled seasonal variation, we exported the modelled daily soil temperature, soil moisture and methane emissions and plotted these to the same x-scale (Figure 4). Here, a negative longer-term correlation could be seen to exist between CH_4 emissions and temperature (Figure 5), with warmer spring/summer temperatures controlling the gradual drop in surface flux for these times. Meanwhile, shorter-term variability in CH_4 flux modelled appears to be linked to changes in soil moisture (driven by climate model input for precipitation) whereby short lived and rapid spikes in soil moisture content lead to spikes in surface fluxes.

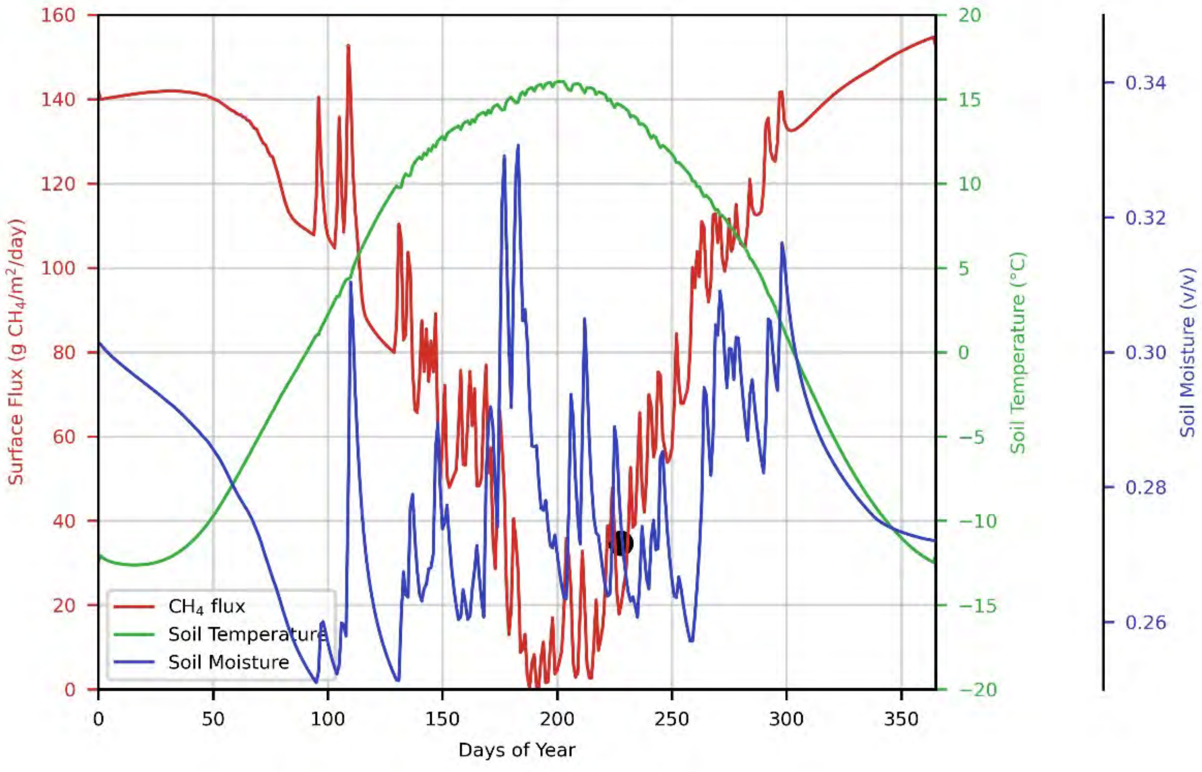


Figure 4: A comparative plot showing the impact of two climate-related inputs (soil temperature and moisture) on the modelled methane emissions (CH_4 flux).

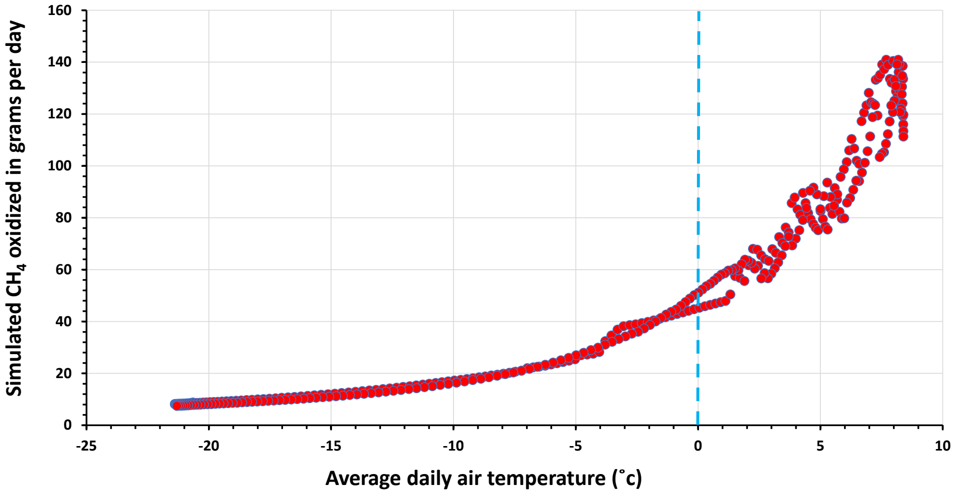


Figure 5: Modelled mass of CH_4 oxidized versus utilized average daily air temperature showing that extent of oxidation is strongly controlled by temperature in the CALMIM model

3.2.Modelling Scenario 2: Impact of Soil Type

Initial models utilized thus far included a soil type specified in CALMIM as silty loam, as was identified as present at wellsite WA22800 (i.e. during Project ES-Wells-2021-02). In order to explore the potential influence of soil composition on CH_4 oxidation extents and emissions a further series of simulations were undertaken varying the soil type with results shown in Figure 6. Here, significant variation in CH_4 emissions are modelled across the four different soil types considered, with this effect being most pronounced during the winter months during which clay soils are modelled to emit more than three times less CH_4 than a sandy soil. However, it is noted that the levels of oxidation are similar across the soil types during the winter months, suggesting this difference within the simulation is due to the inherent physical transport capabilities of the soil, rather than the impact of soil type on oxidising bacteria and their associated reaction kinetics. Conversely, during the summer months, soil type influences surface CH_4 emissions less significantly, but the total amount oxidised varies largely (Figure 6, stippled lines).

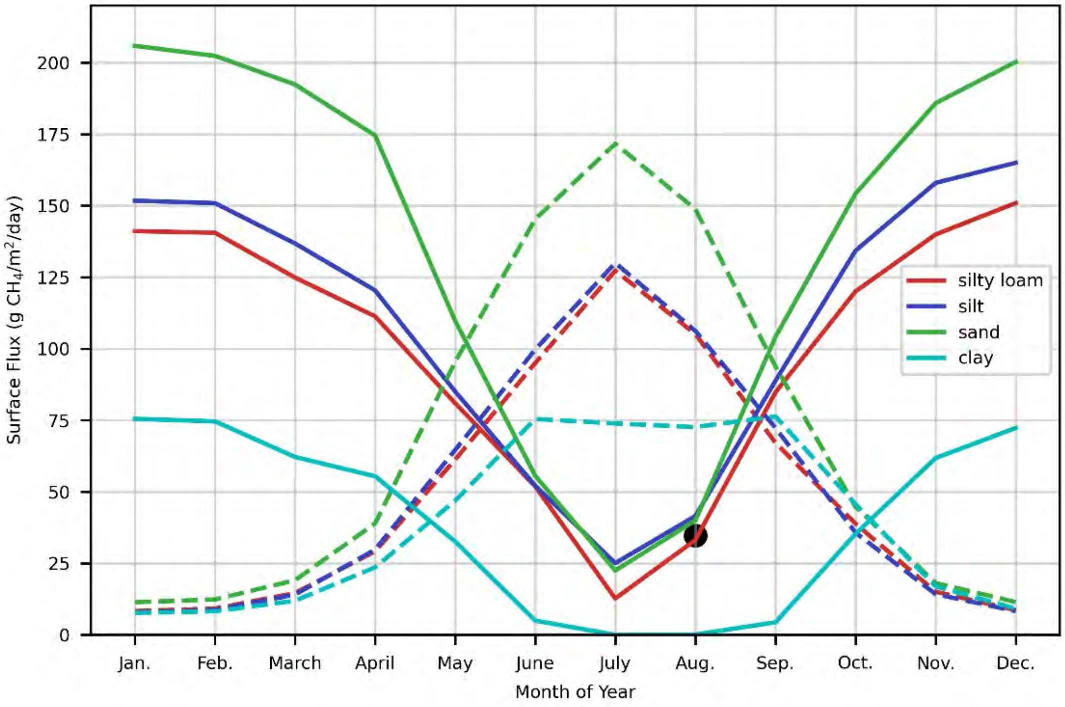


Figure 6: Results from a series of CALMIM simulations to test the sensitivity of the specified soil type on methane emissions. All other parameters except soil type are maintained.

4. Discussion and Conclusions

The following key points result from the modelling exercises conducted using CALMIM as described:

- Methane emissions modelled using CALMIM are driven by climatic conditions (temperature, precipitation), soil properties (type, thickness, maximum oxidation rate) and methane concentration at depth.
- Large swings in air temperature across the year (-15 °C to 15 °C) led our simulations to predict methane emissions that vary by at least a factor of ten. In some of our simulations, very low or no methane emissions were modelled during the summer months but emissions > 100 g/m^2/day were modelled during winter months.
- This suggests that it is vital to incorporate climatic conditions in annual emission estimates. In our case, if we extrapolate our estimated August oxidation rate to a whole year, a total of 12.6 kg/m^2 of CH_4 are predicted to be emitted. By including climatic variability and soil transport properties in CALMIM whereby extent of oxidation is controlled by temperature, we reach an annual value three times larger, at 36.15 kg/m^2.
- As CALMIM does not, to the best of our knowledge, account for frozen ground and snowpack associated with sub-zero temperatures, the very high emissions modelled during the winter months should be treated with an element of caution. It is more likely that emissions will be

lower during the coldest months during which time the frozen uppermost soil might act as a natural confining unit, with methane concentrations building up beneath. As the snowpack and ground melts during early spring, contained methane might be released quickly all at once, resulting in a sharp peak in emissions before stabilising to an emission profile more similar to that which CALMIM predicts.

- It should be noted that our modelling is calibrated to only a single data point (i.e. fluxes measured at the end of August 2023), therefore it is crucial that additional field data is collected, ideally during fall, spring and if possible during the winter months to validate the modelled fluxes and outputs.

Potential for Natural Soils-Based CH_4 Oxidation as a Management or Remedial Strategy for Integrity Compromised wells

Here we conducted research to better understand the rates and extent of CH_4 oxidation in natural soils in BC with a view to use of this process in aiding the management of well integrity failure and fugitive gas leakage. We now integrate the findings and learnings made to comment and explore the potential and limitations of the phenomena to be used as a management approach for integrity compromised wells in British Columbia. The following serve as main discussion points for consideration:

Oxidation of CH_4 in natural soil systems is likely ubiquitous, occurs inherently and is already mitigating emissions at integrity compromised oil and gas wells in BC and elsewhere.

Based on this research project and research cited within it, it is clear that CH_4 oxidation in soils is ubiquitous and readily occurs at potentially significant rates and extents in natural soils around oil and gas wells which are leaking CH_4 whether intended or not. In this case, the process is already acting as a mitigating factor likely at all and any wells where fugitive CH_4 is released into surrounding soils irrespective of whether the concept has been approved or considered as a potential management approach. It is recommended that a greater understanding on this unintended process be attained and there seems no reason why it is not further harnessed or fostered in suitable situations and quantified to understand its contribution to limiting GHG emissions at integrity compromised wells.

Various factors control rates and extents of CH_4 oxidation in natural soils and more work is needed to understand their interplay to attain maximal or optimal conditions. This means the benefits of the process will be variable.

We have described here (through data explored in this study and cited literature) that a range of parameters and properties will influence the overall rates and extents of natural soils based CH_4 oxidation. Clearly there will be locations/settings and times (in terms of seasonal influence) when oxidation is more extensive and occurs at a higher rate. Conversely for certain locations/settings and times the process will be less beneficial, potentially even ceasing to operate or be beneficial at all. In this case, it will be necessary to estimate annual average potential benefits and also to identify locations where characteristics will ensure the greatest potential rates and extents in order to meaningfully asses where and when the process could be used as an authorized mitigation or management strategy for integrity compromised wells. Even though the process might not offer 100% mitigation at all times, the overall average benefits may still outweigh the costs and risks of traditional approaches to managing leaking wells (e.g. full re-entry and re-abandonment) and more consideration is needed to determine if and when such an approach might be the optimal outcome for all stakeholders.

Minimal site set up pre- or post-leakage could enhance and optimize soils based CH_4 Oxidation

From our research and consideration of other research contributions, it is clear that even natural soils can offer a potentially significant ability to mitigate CH_4 leakage with no design or planning. However, it is reasonable to highlight that some simple alterations/augmentations after leakage is discovered or even prior to leakage (for example; perhaps built in to well construction and abandonment procedures) could ensure optimal mitigation by soils based CH_4 oxidation for minimal cost. One key concept to attain optimal soils based CH_4 oxidation is to disperse leaking gases over an appropriate reactive contact area of soils and have an optimal porous media through which gases can flow-through. In this case, after leakage is found (or even pre-emptively during abandonment), a simple, relatively small structure could be created through shallow excavation (e.g. to ~1m depth) with installation of a thin gas distribution sand/gravel layer (e.g. 0.2 m thick) backfilled on top with natural soils and potentially some amendments depending on composition (e.g. sand, compost). This will ensure any leaking gas is dispersed over a wider area/volume of soils and moves moderately though the reactive layer. The structure might only need to be in the order of 20 – 80 m^2 in area (i.e. a square of 5 to 9 m in lengths) to offer potential for mitigation of leakage up to 5 to 10 m^3/day (as indicated by this study) of a varying total extent depending on the seasons. Such a structure could achieve greater extents and rates than described in natural soil structures and could have very low costs (in the order of ~$10 - 20k) A more detailed design and trial program would be needed to scope out such an option more robustly and is recommended to be explored/considered by all stakeholders. A concept for a minimally engineered soil system to optimize CH_4 oxidation is shown below in Figure 1.

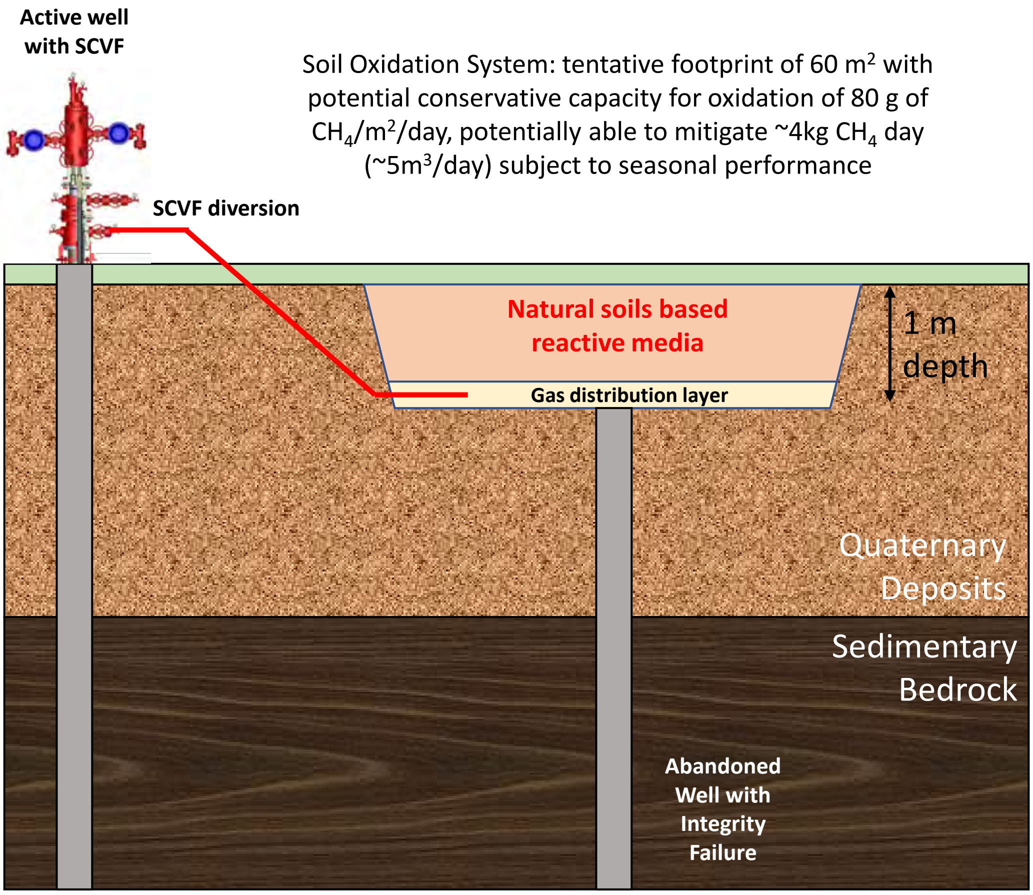


Figure 1: Concept sketch for a simplistic Soil Oxidation System (SOS) to mitigate active and abandoned well CH_4 leakage with minimal engineering/site preparation with conservative sizing and capacity estimates. The SOS could be constructed pre- or post-leakage detection and sized according to load with the reactive media formed primarily by natural site soils with augmentations depending on composition to optimize gas transport and reactive contact time. More research is needed to robustly design such a system and further evaluate the process the soil oxidation process and optimize it.

Natural soils-based CH_4 oxidation could be used to mitigate SCVF during a wells active life.

In this project, we have focused on the leakage of CH_4 from abandoned wells. However, for minimal effort it could be possible to harness the process to partially (or even fully) mitigate SCVF for wells during their active life. Here, instead of emitting CH_4 directly to the atmosphere via the SCV during active life (as is common practice), soil diversion to shallow depths with minimal engineering and site preparation could lead to significant benefits in terms of GHG emissions reductions. It is recommended to further consider and evaluate a potential approach such as this whereby potentially significant reductions in CH_4 emissions could easily be achieved. In particular wells with <10m^3/day of SCVF which are not typically subject to remedial action and form the most common flow rates in BC (i.e. comprising 80% of reported SCVF according to Wisen *et al.*[6]) could be almost fully mitigated by soils-based oxidation leading to potentially valuable GHG emission reductions.

A consortium of micro-organisms drive soils based CH_4 oxidation and more understanding on their functioning and interplay is needed to further optimize and potentially enhance the process.

We showed clearly that no single microorganism drives soils based CH_4 oxidation, with a consortia acting together to drive the process in a multi-sept manner more likely. In this case, further understanding on the exact taxa and the mechanisms/steps involved could lead to identification of further ways to optimize and enhance the process in natural soils. It is recommended that further research is conducted that provides more microbial detail on the process such that it can be more effectively harnessed.

Overall, this project has attained and reviewed data that shows natural soils based CH_4 oxidation has potential to be employed as a low-cost, low-tech management strategy for reducing or even fully mitigating CH_4 emissions from integrity compromised oil and gas wells. However, more understanding and consideration of how to optimize and appraise the process is needed before it can be deployed. It is recommended that more research to explore this approach is commissioned that can further scrutinize and explore the process and evaluate where and when it could be used as well as how to optimize the process.

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