



Measuring methane emissions from oil and gas platforms in the North Sea

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Abstract. Recent studies suggest oil and natural gas production facilities in North America may be underestimating methane (CH₄) emissions during extraction. This, coupled with unusually high CH₄ mole fractions observed at coastal sites during onshore winds in the UK, suggests CH₄ emissions from oil and gas extraction activities in the North Sea could be higher than previously reported. To investigate if these coastal CH₄ enhancements could have come from oil and gas production platforms, we use near-source measurement techniques to estimate CH₄ emissions from eight oil and gas production platforms in the North Sea. We estimate the mean CH₄ emission from the eight platforms to be 10.1 g CH₄ s⁻¹, with a range of 1.1 to 25.0 g CH₄ s⁻¹. When matched to production records, individual platforms lose between 0.01% and 1.58% of gas production with an average loss of 0.61% of gas production. However, when the measured platforms are considered collectively, i.e. when the total measured emission is compared to total production of the platforms measured, the CH₄ loss is estimated at 0.27% of gas production. These calculated ranges are at least double the most recently reported loss rates for these platforms, which are currently estimated at 0.13% of gas production. In fact, the vast majority of reported emissions are due to gas flaring and offshore oil loading, neither of which was taking place at the time of these measurements. If emissions measured here resulted from leakage during normal operation, they represent significant additional emissions (at least 0.27% of production) above previous estimates of CH₄ leakage from off-shore oil and gas production platforms. These emissions are not explicitly included in UK emission inventories. Further research to determine CH₄ leakage from all operations occurring at off-shore oil and gas platforms, and how to include them in national emission inventories, is needed.

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1 Introduction

Between 2012 and 2015 unusually high methane (CH_4) enhancements, of up to 400 ppbv above background, were observed at the University of East Anglia's Weybourne Atmospheric Observatory (WAO; 52.95 °N, 1.14 °E) during periods of northerly on-shore winds and high pressures (Connors, 2015; Staunton-Sykes, 2016; Connors et al., 2018). These elevated enhancements were unexpected as the air came from the open ocean. However, a potential source of these CH_4 enhancements is leakage from offshore oil and gas production platforms 80 km away from WAO in the North Sea (OSPAR, 2018; Fig. 1).

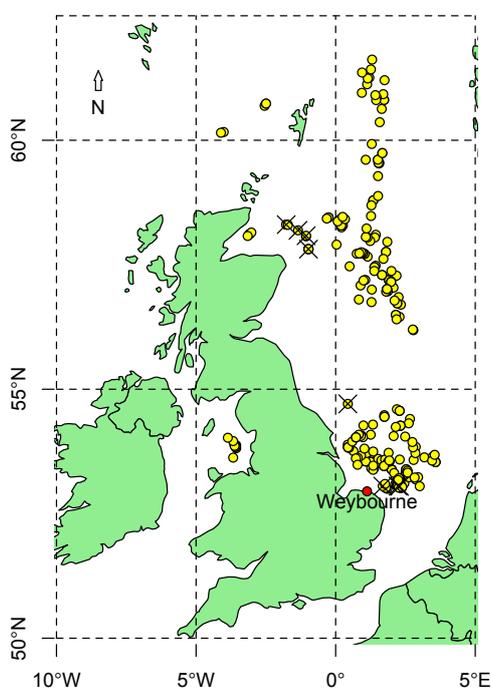


Figure 1 Map of the North Sea showing the locations of all UK offshore oil/gas platforms (the filled yellow circles) and the eight platforms measured by this study (black crosses) (Source: OSPAR, 2012).

- 10 In 2015 the UK extracted about 32 Tg of natural gas from the North Sea (OGA, 2018). During this time, a loss of 40 Gg CH_4 (0.13 % of natural gas production) was reported by the UK Government Department for Business, Energy & Industrial Strategy (BEIS), mainly through venting (24 Gg $\text{CH}_4 \text{ yr}^{-1}$) and flaring (12 Gg $\text{CH}_4 \text{ yr}^{-1}$) activities (BEIS, 2018). However, recent measurement studies suggest public inventories underestimate CH_4 emissions and that emissions from the offshore platforms could be higher (Zavala-Araiza et al., 2015, Schwietzke et al., 2017).
- 15 Land-based measurements in West Virginia and Colorado, USA, estimate on-shore oil and gas extraction activities lose between 0.1 and 10 % of CH_4 produced (Petron et al., 2012, Omara et al., 2016; Schwietzke et al., 2017; Englander et al., 2018; Riddick et al., 2019). There are no direct measurements of CH_4 emissions from offshore oil and gas production. However, qualitative studies identify offshore oil and gas operations as large, yet uncertain, emission sources (Nara et al.,



2015; Allen et al., 2016). Numerous productive offshore fields exist across the globe, including the Gulf of Mexico, California, Brazil, and Newfoundland and Nova Scotia, making this an area worthy of careful measurements and analysis.

Several sources of CH₄ from off-shore production platforms are explicitly identified in the BEIS emissions inventory and include: combustion activities such as gas flaring, offshore oil loading and venting directly to the atmosphere (BEIS, 2018).

5 However, venting of casing gas and incomplete combustion of gas at the flare stack are not explicitly identified by the BEIS, and these were found to be the most likely sources of unreported emissions at onshore oil and gas extraction sites in Alberta (Johnson et al., 2017).

The CH₄ emissions reported by the BEIS from the UK offshore oil and gas sector are primarily based on data self-reported by the oil and gas operators. Emissions data are calculated by the operator via a combination of emission monitoring and emission
10 factors. Technical guidance on the emission factor based calculations is available through the UK Government's Department of Energy and Climate Change (DECC EEMS, 2008), but no guidelines have been published for direct monitoring systems. The main shortcoming of using emission factors is that the total emission can be underestimated if not all sources are identified. Overall, the approaches used by industry are potentially underestimating emission factor calculations and unregulated direct measurements, both of which could lead to underestimates of CH₄ emissions from offshore installations.

15 In this study we investigate whether CH₄ emissions from offshore oil and gas installations in the North Sea differ from those currently reported by the BEIS. To investigate the loss of CH₄ from offshore oil and gas installations we use two approaches; 1) determine whether the source of CH₄ enhancements at WAO could be from oil and gas production platforms; and 2) estimate an average CH₄ loss from offshore installations by making direct measurements of CH₄ emissions from off-shore production
20 platforms in the North Sea. To our knowledge, this is the first time that CH₄ emissions from off-shore oil and gas production platforms have been quantified.

2 Methods

2.1 Land based observations

In order to investigate the source of the CH₄ enhancements observed at WAO during a northerly wind event, atmospheric air samples were collected at Weybourne, UK (Fig. 1) between 10:00 and 13:00 on 13th October 2015. Air samples were pumped
25 periodically into 1 L Tedlar bags 4 m above sea-level at WAO. The Tedlar bags were partially filled and then totally expunged prior to the collection of each air sample to minimize contamination. Carbon isotope signatures were later measured from the air samples, at Royal Holloway, University of London, using a modified commercially available continuous flow gas chromatography/isotope ratio mass spectrometer (CF-GC/IRMS) allowing high precision ($1\sigma = 0.05\%$) carbon isotopic analysis of CH₄ at ambient mole fractions, with set-up and calibration as described in Fisher et al. (2006).



2.2 Boat based observations: Production platforms

As of March 2017, there were 260 operational offshore oil and gas platforms run by 27 different operators in UK waters (OGA, 2017). The oldest platforms have been producing since 1967. These platforms are found between 30 and 500 km from the UK mainland, with the majority of platforms located to the East of the UK in the North Sea (Fig. 1; OSPAR, 2018). To investigate possible emissions from these platforms, sea-level CH₄ mole fractions were measured around eight oil and gas platforms between the 6th June and 25th August 2017. Measurements from boats of CH₄ emissions from individual production platforms were made during normal operation (i.e. pilot light on the flare stack is lit but no flaring or offshore oil loading is taking place). To determine if flaring or offshore oil loading was happening a visual inspection was made of the installation. This level of targeting was not achievable using land-based measurements made 50 km away at WAO. The previously published emissions from the platforms are reported to be almost entirely due to flaring (83%) and offshore oil loading (17%), where the emissions were generated using emissions factors (Brown et al., 2017; BEIS, 2018).

The oil and gas platforms measured here were selected at random, constrained only by the need for accessibility. Fishing boats were chosen as the measurement platforms because of budgeting and availability constraints; Maritime and Coastguard Agency regulations for the available vessels (MCA category 2) meant that the platforms measured had to be less than 60 miles from a safe haven. Four of the eight platforms chosen primarily produced natural gas, while the remaining four produced oil and gas. Methane mole fractions, latitude, longitude, and meteorological data were collected as the boat travelled upwind and downwind of the platforms.

2.2.1 Methane mole fraction measurements – Los Gatos UGGA

The Los Gatos Research Ultra-portable Greenhouse Gas Analyzer (UGGA; www.lgrinc.com) is a laser absorption spectrometer that measures CH₄ mole fractions in air using off-axis integrated cavity output spectroscopy (Paul et al., 2001). The UGGA reports CH₄ mole fractions every second, with a stated precision of < 2 ppb (1 σ @ 1 Hz) over an operating range of 0.1 to 100 ppm. Calibration of the UGGA was done before and after deployment using low (1.93 ppm CH₄), target (2.03 ppm CH₄) and high (2.74 ppm CH₄) mole fraction gases calibrated on the WMO scale. Measurements were taken between the edge of the exclusion zone (500 m from the platform; HSE, 2018) and 1.5 km horizontal distance from the platforms. The inlet line was attached to a mast 2.5 m above sea level, to avoid contamination from the boat's exhaust, and protected from water incursion using an aluminium funnel and the air was filtered using a 2 μ m filter. Background CH₄ mole fractions were measured while the boat was upwind of the production platform.

2.2.2 Meteorological data

In-situ meteorological data were collected using a wireless weather station (Maplin, UK) attached to a mast 2 m above sea level. Meteorological data were sampled and recorded at one-minute intervals and included: wind speed (u , m s⁻¹), wind direction (WD , ° to North), air temperature at 2 m (T_a , K), relative humidity (RH , %), rain rate (R , mm hr⁻¹), irradiance (I , W



m⁻²) and air pressure (P , Pa). The wind speed used in the emission modelling was corrected for emission height using a wind profile power law (Touma, 1977; Hsu et al., 1994).

2.2.3 Gaussian Plume Model

The Gaussian Plume (GP) model used in this study describes the mole fraction of a gas as a function of distance downwind from a point source (Seinfeld and Pandis, 2006). As a gas is emitted, it is entrained in the prevailing ambient air flow and disperses in the y and z directions (relative to a mean horizontal flow in the x direction) with time, forming a cone. The gas is considered to be well mixed within the volume of the cone, such that the mole fraction of the gas as a function of distance downwind depends on the emission flux at the source, the advective wind speed (u , m s⁻¹), and the rate of dispersion. The mole fraction of the gas (X , µg m⁻³), at any point x metres downwind of the source, y metres laterally from the centre line of the plume, and z metres above ground level can be calculated (Eq. 1) using the source strength (Q , g s⁻¹), the height of the source (h_s , m) and the air stability. The standard deviation of the lateral (σ_y , m) and vertical (σ_z , m) mixing ratio distributions are calculated from the stability class of the air (Pasquill, 1974). The Gaussian plume approach assumes that the vertical eddy diffusivity and wind speed are constant and there is total reflection of CH₄ at the surface and this method has been used to measure CH₄ emissions from

$$X(x, y, z) = \frac{Q}{2\pi u \sigma_y \sigma_z} e^{\frac{-y^2}{(2\sigma_y)^2}} \left(e^{\frac{-(z-h_s)^2}{(2\sigma_z)^2}} + e^{\frac{-(z+h_s)^2}{(2\sigma_z)^2}} \right) \quad (1)$$

2.2.4 Gaussian Plume model parameterization

A GP approach was used to infer the CH₄ emissions from the oil and gas platforms using the CH₄ mole fraction data collected downwind. We used measurements for the mole fraction, and rearranging Equation (1) solved for the source term Q . Data used as input to the GP model are: wind speed, wind direction, temperature, CH₄ mole fraction at 2 m, background CH₄ mole fraction and the Pasquill-Gifford stability class (PGSC). The PGSC are estimated from wind speed and irradiance data (Seinfeld and Pandis, 2006), as measured by the meteorological station on the boat.

The height of the working deck and the height of the flare stack at each of the platforms were determined using the database of platform characteristics. Emissions were calculated for three scenarios: 1) The source was at the height of the working deck level, 2) The source was at the top of the flare stack and 3) A best-estimate (see Section 3.4 below), where the emission was between the working deck and the flare.

2.2.5 Data sources

The UK Department for Business, Energy & Industrial Strategy (BEIS) keeps the Environmental and Emissions Monitoring System (EEMS) which is the environmental database of the UK oil and gas industry. Methane emission data are uploaded to

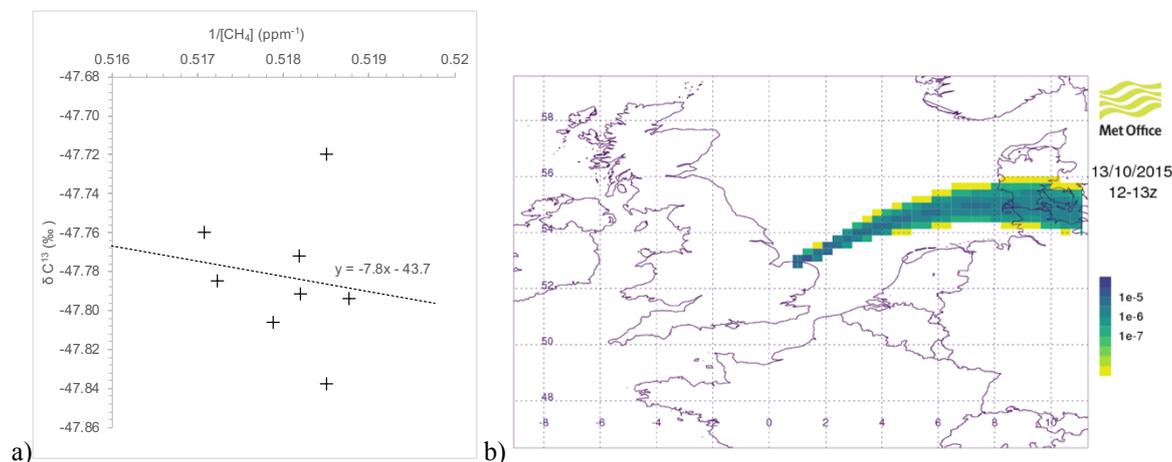


5 this by industry partners. These data form the basis for emissions reported under category 1B2 within the National Atmospheric Emissions Inventory (NAEI; BEIS, 2018). For details of how this data is incorporated into the NAEI, see Brown et al. (2017). The most recent point-source emission database from the NAEI available at the time of writing was for the year 2015. Individual platform production data for both 2015 and 2017 were taken from the Petroleum Production Reporting system published by the UK Oil and Gas Authority (OGA, 2018).

3 Results

3.1 Methane sources during northerly wind events

10 Measurements of the mole fraction and the carbon isotopic signature of the CH₄ contained within the Tedlar bag IR samples were measured at RHUL and identified the CH₄ source to have a carbon isotopic signature of -43.7 ‰ (Fig. 2a) for the air collected during the northerly wind event on 13th October 2015 (Fig. 2b). This falls within reported carbon isotopic range for Southern North Sea gas, -34.5 to -45.3 ‰ (Hitchman et al., 1989) and implicates Southern North Sea gas operations as a plausible source for the enhancements observed during northerly wind events at WAO.



15 **Figure 2a)** Keeling plot of atmospheric air samples collected at Weybourne, UK during the northerly wind event between 10:00 and 13:00 on 13th October 2015 showing a carbon isotopic signature of -43.7 ‰. **b)** 2-day back-trajectory of winds arriving at WAO between 12:00 and 13:00 on 13th October 2015 shows the surface areas impacting the observation point. The scale shows integrated concentration, g s⁻³, (concentration of tracer particle integrated over the back trajectory run duration) with blue representing the highest integrated concentration.

3.2 Methane mole fractions around the platforms

20 Measurements at sea-level indicate CH₄ mole fraction enhancement can be measured at all of the production platforms, when the upwind CH₄ mole fractions ($[CH_4]_{bgd}$, ppb) are compared to the downwind mole fractions ($[CH_4]$, ppb; Table 1). The largest enhancement, 370 ppb, was observed downwind of Platform #6 on the 24th August, the lowest enhancement, 11 ppb,



was observed downwind of platform #8 on the 25th August. The mean CH₄ enhancement downwind of the 8 platforms was 112 ppb.

5 **Table 1 Meteorological, position and mole fraction data taken during the boat-based measurement campaign around production platforms in the North Sea between 6th June and 25th August 2017. Emissions were calculated for three scenarios: 1. Lower estimate: The source was at the working deck level, 2. Upper estimate: The source was the top of the flare stack and 3. A best-estimate, where the emission was between the working deck and the flare. Numbers in parentheses indicate lower and upper bounds for the indicated variables.**

ID #	Date	Type	Height (m)	Distance (m)	Downwind [CH ₄] (ppb)	Emission (g s ⁻¹)	Platform Gas Production (g s ⁻¹)	Loss (%)
1	Jun 6	Gas	50 (40, 60)	2000	1980	6.6 (6.4, 6.9)	N/A	
2	Jun 6	Gas	50 (40, 60)	2000	1981	6.7 (6.5, 7.0)	N/A	
3	Jul 5	Gas	50 (40, 60)	2000	1950	3.9 (3.8, 4.0)	672	0.58
4	Aug 16	Gas	70 (50, 90)	600	2090	10.3 (4.7, 29.2)	15,230	0.07
5	Aug 24	Oil/Gas	70 (50, 90)	600	2290	25.0 (11.4, 71.1)	1,585	1.58
6	Aug 24	Oil/Gas	70 (50, 90)	550	2232	23.7 (9.5, 80.5)	1,845	1.28
7	Aug 25	Oil/Gas	70 (50, 90)	600	1979	3.1 (1.4, 8.9)	2,952	0.11
8	Aug 25	Oil/Gas	70 (50, 90)	550	1955	1.1 (0.5, 3.8)	8,047	0.01
					Average	10.1 (5.5, 26.4)		0.61 (0.31, 1.66)
					Total	80.4 (44.2, 211.4)	30,333	

3.3 Source of leaks

10 Even though the production platforms measured in this campaign were not actively flaring, (i.e. burning gas as a pressure relief system during oil extraction), the pilot light on the top of the flare stack was actively burning gas on most of the platforms. As an example, Fig. 3 shows an example of minute-averaged CH₄ mole fraction measurements made upwind and downwind of a production platform on the 24th August. This example was chosen as it was the only platform that had an offset flare stack (i.e. not centred in the platform), which better demonstrates the potential source of the emission. Fig. 3 indicates background (1.92 ppm) CH₄ mole fractions around the platform except for an obvious increase in mole fraction downwind of the flare stack.

15 The width of the plume (< 200 m) suggests a compact CH₄ source. This could be associated with incomplete combustion of natural gas feeding the pilot light at the top of the platform, or it could be associated with gas emission at the working deck level.

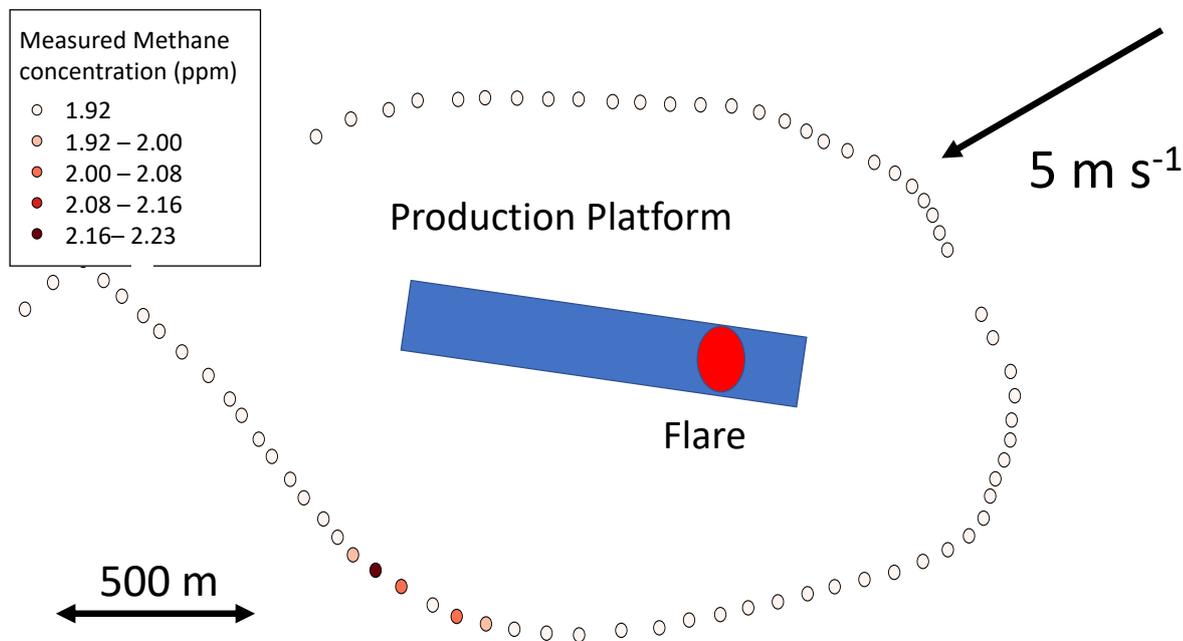


Figure 3 Minute-averaged CH_4 mole fraction measurements made upwind and downwind of production platform, ID # 5, on the 24th of August.

3.4 Estimating methane emissions

- 5 The highest emission of 25.0 g s^{-1} (with lower and upper bounds estimated to be 11.4 and 71.1 g s^{-1} , respectively) was observed at platform ID # 5 on the 24th August, while the lowest emission, 1.1 (0.5 , 3.8) g s^{-1} , was observed on the 25th August at platform ID # 8. The mean CH_4 emission from the eight platforms was 10.1 (5.5 , 26.4) g s^{-1} . When normalized against natural gas production data (OGA, 2018), the highest CH_4 loss was 1.58% of production at platform ID # 5 while the lowest loss was 0.01% of production at platform ID # 8. The average CH_4 loss is estimated at 0.61% of production for the six platforms for which we could obtain production data, with a range of 0.31% to 1.66% depending on assumed emission height.
- 10 When weighted by production, i.e. the collective emission from the platforms measured (80 g s^{-1} ; Table 1) as a fraction of the collective production of the measured platforms ($30,333 \text{ g s}^{-1}$; Table 1), the average loss from all platforms measured was 0.27% of the total production, with a range of 0.15% to 0.70%. The production data for the platforms measured on the 6th June and 5th July were not available.
- 15 For comparison, we have also calculated the reported loss rates for 2015 using the most recent NAEI emissions data (Brown et al., 2017; BEIS, 2018). We find the average reported loss rate was 0.76% for the six platforms where production data was available, with a production-weighted average of 0.20%. These values are within the ranges calculated above for our measured emissions. However, this apparent consistency is highly dependent on assumptions regarding the source-type of the measured



emissions as the NAEI emissions also account for CH₄ emissions from flaring and offshore oil loading activities; this is discussed further in section 4.

3.5 Uncertainty in emission estimates

The main uncertainty using the Gaussian plume approach in this study is in estimating the height of emission. Our best emission estimate suggests the emission height is half way between the platform and the flare and is taken as the average of the emissions calculated at these heights. Uncertainty in the CH₄ release point results in three average CH₄ emissions from the eight platforms of: 5.5 g s⁻¹, where the emissions all come from the production level, 26.4 g s⁻¹, where the emissions are from the flare and 10.1 g s⁻¹ a best-estimate as an average between the two heights.

In addition to uncertainty in emission heights, there is also uncertainty in the emission estimate caused by the GP model. Scenarios were run using the GP approach to reflect variability in instrument precision, wind speed, temperature and the PGSC. Instrument precision and air temperature had little effect on the emission estimate, both ± 1 %. The calculation of the PGSC was the largest source of uncertainty as the PGSC was calculated from the atmospheric conditions at sea-level. However, this may not fully represent the stability of a gas coming from a production platform. As an estimate we suggest the atmospheric stability of this CH₄ could be one PGSC less stable and the resulting variability in CH₄ emission could be ± 54 %. The overall uncertainty in CH₄ emission, calculated as the root of the sum of the individual uncertainties squared, is estimated to be ± 54 %.

4 Discussion

Our measurements of CH₄ emissions from off-shore oil platforms are insufficient to explain the large CH₄ enhancements observed at WAO during periods of onshore winds (i.e. enhancements of 400 ppb above background measured 80 km from the platforms). However, isotopic data collected at WAO indicate that the CH₄ is from a thermogenic source. The enhancements could be from operational activities not observed during our measurement campaigns, such as venting or flaring, where larger amounts of CH₄ are emitted. However, the air history map suggests thermogenic CH₄ pollution from continental European (Fig. 2b) is also a plausible source as the carbon isotopic signature of continental gas and CH₄ from coal is similar to North Sea gas (Dlugokencky et al., 2011). Cain et al. (2017) describe similar problems of continental pollution when measuring enhancements downwind of North Sea gas platforms. The compounding effects of pollution from either the UK or continental Europe suggest land-based observation of North Sea platform emissions may not be effective and near-source measurement campaigns targeting individual platforms are necessary to characterise their emissions.

The near-source measurements presented in this study estimate an average CH₄ emission from offshore oil and gas installations of 873 kg CH₄ day⁻¹, with individual platform's CH₄ emissions ranging from 95 to 2,160 kg CH₄ day⁻¹. Matching production data to our measurements we estimate two metrics 1) an average loss of CH₄ from the six platforms unweighted by production at 0.61% (range 0.31 to 1.66%) and 2) the summative loss of CH₄ weighted by total production at 0.27% (range 0.15 to 0.70%).



These results show, of the platforms measured, those producing more gas leak proportionally less of what they produce. Also, the two higher emitting platforms (IDs 5 and 6) include floating production storage and offloading vessels (FPSOs); we find these to have much larger loss rates than the three fixed platforms (IDs 4, 7 and 8). However, we also acknowledge the sample size is small and the six platforms may not be representative of the overall performance of platforms in the North Sea.

5 Using 2015 emission factor based NAEI emissions, the same metrics for these platforms are within the ranges calculated in this study, i.e. an average loss rate of 0.76% and a production-weighted average loss of 0.20%, and also show larger losses come from lower producing platforms. If the measurements presented here represent continuous emissions at a constant rate from these platforms, we might conclude that the measured emission are consistent with the values reported by the NAEI. However, the NAEI also states the main source of emission for each installation and the emissions from the six platforms are

10 almost entirely due to flaring (83%) and offshore oil loading (17%). Typically, these activities are not continuous on North Sea platforms; consequently, emission rates are likely to be much higher at certain times than others. As flaring and oil loading did not coincide with our measurement campaign, the measured emissions presented here represent leakage only and do not account for intermittent emissions due to venting, flaring or oil loading activities. This suggests a potentially large source of CH₄ emission has not been quantified or accounted for in the national CH₄ emission inventory. Further work is needed to

15 measure more platforms over a longer time frame to better understand the role of ambient leakage, venting, flaring and oil loading to investigate whether large emissions, as seen at WAO, are possible. This would be made easier if records detailing when venting, flaring and oil loading have occurred are made publicly available.

The continuous leakage of CH₄ from offshore production platforms observed here is consistent with observations of similar onshore operations (Omara et al., 2016; Riddick et al., 2019). Ambient leakage is not unexpected as these off-shore production

20 platforms are located in the inhospitable conditions of the North Sea, where wind speeds regularly exceed hurricane force and waves can reach the working deck of the production platform. However, it is surprising that ambient leakage has not been explicitly factored into the UK national emissions inventory, which relies solely on operators self-reporting emissions that are calculated using bottom-up methods. Without direct measurement, operators are unaware of small emissions that may occur during normal operation.

25 The small amount of CH₄ lost as ambient leakage measured here may not be economically important, but the loss of 0.27% of production (the production-weighted average loss) corresponds to a global emission of 1.4 Tg CH₄ yr⁻¹ (IEA, 2018). Currently, the global CH₄ emission from the oil and gas sector is estimated at 1.6 Tg CH₄ yr⁻¹, based on a 0.32% baseline of the Oil and Gas Climate Initiative (OGCI, 2018). This estimate represents data from 13 of the largest oil and gas producers and accounts for the total up-stream CH₄ emissions from all operated gas and oil assets and includes flaring, venting and offshore oil loading.

30 When emission from ambient leakage presented by this study (1.4 Tg CH₄ yr⁻¹) is added to the current estimate from flaring, venting and offshore oil loading (1.6 Tg CH₄ yr⁻¹) the total emission is almost doubled. This study highlights the shortcomings of using emission factors which rely on *a-priori* knowledge of the source, in contrast direct measurement account for all emission and present a better method for estimating total emissions. In conclusion, we suggest that additional *in-situ* measurements of offshore oil and gas production platform operations (the Gulf of Mexico, California, Brazil, and



Newfoundland) be conducted to better inform leakage estimates and that these measurements be used to improve the UK and global CH₄ emission inventories.

Author contributions

For the isotopic methane experiment at WAO, J. Staunton-Sykes, S. N. Riddick and N. R. P. Harris designed the experiment, J. Staunton-Sykes and G. L. Forster prepared equipment and calibrated the instruments, J. Staunton-Sykes and G. L. Forster carried out the measurements and J. Staunton-Sykes, D. Lowry, E. G. Nisbet analysed the samples. For the offshore measurements, S. N. Riddick designed the experiment, S. N. Riddick, G. Allen, J. Pitt prepared equipment and calibrated the instruments and S. N. Riddick carried out the measurements. A. J. Manning performed the inversion simulations. D. L. Mauzerall and M. Celia were project leads and gave scientific oversight and guidance throughout the planning, implementation, collection, and analysis of the data. S. N. Riddick, D. L. Mauzerall, M. Celia, M. Kang and N. R. P. Harris prepared the manuscript with contributions from all co-authors.

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15