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Occurrence and fate of methane leakage from cut and buried abandoned gas wells in the Netherlands

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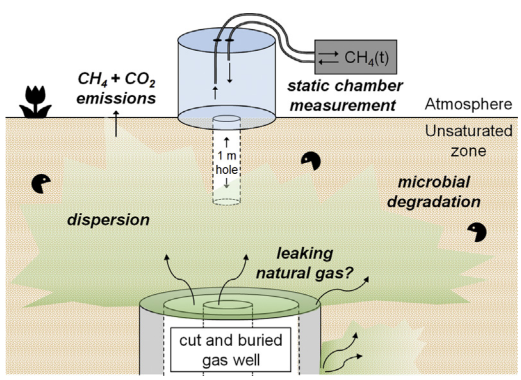
HIGHLIGHTS

- Monitoring for gas leakage at cut and buried wells is complicated by several factors.
- Leakage was detected at 1 of 29 cut and buried abandoned wells in the Netherlands.
- Dispersion and oxidation in soils can prevent detection by surficial measurements.
- Subsurface and isotopic measurements needed to confirm and quantify methane leakage.
- Cut and buried wells may constitute an explosion hazard that needs to be considered.

GRAPHICAL ABSTRACT



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ABSTRACT

Methane leakage caused by well integrity failure was assessed at 28 abandoned gas wells and 1 oil well in the Netherlands, which have been plugged, cut and buried to below the ground surface (≥3 mbgl). At each location, methane concentrations were thoroughly scanned at the surface. A static chamber setup was used to measure methane flow rates from the surface as well as from 1 m deep holes drilled using a hand auger. An anomalously high flow rate from 1 m depth combined with isotopic confirmation of a thermogenic origin revealed ongoing leakage at 1 of the 29 wells (3.4%), that had gone undetected by surficial measurements. Gas fluxes at the other sites were due to shallow production of biogenic methane. Detailed investigation at the leaking well (MON-02), consisting of 28 flux measurements conducted in a 2 × 2 m grid from holes drilled to 1 and 2 m depth, showed that flux magnitude was spatially heterogeneous and consistently larger at 2 m depth compared to 1 m. Isotopic evidence revealed oxidation accounted for roughly 25% of the decrease in flux towards the surface. The estimated total flux from the well (443 g CH_4 hr^{-1}) was calculated by extrapolation of the individual flow rate measurements at 2 m depth and should be considered an indicative value as the validity of the estimate using our approach requires confirmation by modelling and/or experimental studies. Together, our findings show that total methane emissions from leaking gas wells in the Netherlands are likely negligible compared to other sources of anthropogenic methane emissions (e.g. <1% of emissions from the Dutch energy sector). Furthermore, subsurface measurements greatly improve the likelihood of detecting leakage at buried abandoned wells and are therefore essential to accurately assess their greenhouse gas emissions and explosion hazards.

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1. Introduction

Leakage of natural gas from oil and gas wells has been identified as an environmental hazard for several decades (Dusseault et al., 2000; Erno and Schmitz, 1996; Harrison, 1983). Not only does leakage from oil and gas infrastructure appear to contribute significantly to rising anthropogenic methane emissions and the associated greenhouse effect (Miller et al., 2013; Rice et al., 2016), gas migrating into surrounding geological strata adversely impacts groundwater in aquifers overlying oil and gas reservoirs (Harrison, 1983; Kelly et al., 1985) and may pose an explosion hazard (Chilingar and Endres, 2005). In recent years, elevated levels of thermogenic methane in groundwater have also been attributed to wellbore leakage from newer, unconventional oil and gas wells (Darrah et al., 2014; Sherwood et al., 2016). Although water containing methane is not considered a health hazard (Vidic et al., 2013), the introduction of methane in a groundwater system may result in changing redox conditions that in turn may lead to water quality changes, such as increased sulfide concentrations (Gorody, 2012) or the mobilization of trace elements (Cahill et al., 2017).

Recent conservative estimates suggest that at least four million onshore hydrocarbon wells have been drilled worldwide (Davies et al., 2014). A well may leak if a conduit for fluid migration exists from the wellbore system to its surroundings, a situation that is referred to as well integrity failure (King and King, 2013). Whether leakage to outside the well occurs depends on the presence of a driving force. This explains why buoyancy driven gas leakage is a more common observation than leakage of liquids (Davies et al., 2014) and that the leakage risk appears to be higher for gas wells than for oil wells (King and King, 2013). Other attributes that play a role in determining leakage frequency are differences in well type (conventional versus unconventional), geographic location, well age (Ingraffea et al., 2014), well depth (Watson and Bachu, 2009), and plugging status (Kang et al., 2016). Hence, observed failure frequencies vary greatly between different basins, with reported values ranging from 1.9% to up to 75% (Davies et al., 2014).

In addition to these well attributes, observed leakage frequencies may also differ as a result of the monitoring method used. Detection and quantification of gas leakage caused by well integrity failure is typically assessed at the wellhead using measurements of either sustained casing pressure (SCP) (Brufatto et al., 2003; Lackey et al., 2017), surface casing vent flow (SCVF) (Dusseault et al., 2014; Watson and Bachu, 2009) or visual observation of leaking gas bubbles. Furthermore, gas leakage from oil and gas wells has also been assessed by measuring the concentration and isotopic composition of dissolved gasses in nearby groundwater wells (e.g. Osborn et al., 2011; Van Stempvoort and Jaworski, 1995). However, the ability to detect leakage using groundwater monitoring strongly depends on the distance between the leakage point and the groundwater sampling location (Jackson et al., 2013; Molofsky et al., 2011), the direction and velocity of groundwater flow and the occurrence of microbial methane oxidation (Schout et al., 2017; Van Stempvoort et al., 2005).

At abandoned oil and gas wells, monitoring options are often limited by the lack of an intact wellhead. With the number of abandoned wells growing rapidly (Dusseault et al., 2014), effective monitoring at these wells will be crucial to identify and remedy methane leakage. At 88 abandoned oil and gas wells in Pennsylvania, USA, methane flow could be measured by sealing a chamber over the remainder of abandoned wellbore systems protruding from the surface (Kang et al., 2016). 90% of these wells were a positive source of methane, with gas wells and unplugged wells emitting relatively large amounts of methane compared to oil wells and plugged wells. Besides plugging, some oil and gas jurisdictions require that wells are cut to below the ground surface and buried as the final step in the abandonment process (Davies et al., 2014). At such wells, detecting and quantifying a gas leak at surface is complicated as it originates from an uncertain depth below the surface.

Methane flow rates at 138 abandoned wells from four US states were measured by Townsend-Small et al. (2016), including a subset of wells that had been cut and buried. To measure leakage at these locations, the surface above the well sites was first scanned using mobile methane detection instruments. When anomalous concentrations were detected, methane fluxes at the soil-atmosphere interface were measured using flux chamber methodology and gas samples were collected for source identification. Overall, the study showed that 42% of unplugged wells compared to only 0.8% of plugged wells were a positive source of methane. Boothroyd et al. (2016) measured methane concentrations at the soil-atmosphere interface above 102 plugged, cut and buried wells that had been decommissioned according to best practice in the UK. By comparing the measured concentrations to that at nearby control locations, ongoing leakage caused by well integrity failure was concluded for 30% of these wells. However, the wells being the actual source of the methane were not corroborated by gas sampling and isotopic analysis. In addition, gas leaking from cut and buried wells has to migrate upward through the vadose zone by diffusion and buoyancy, where it is dispersed and may be partially oxidized before it can be detected at the surface (Erno and Schmitz, 1996). Furthermore, preferential flow paths may cause leaking gas to emerge offset with a considerable distance (>10 m) from a leaking wellbore (Forde et al., 2019).

Overall, both the environmental hazard of gas leakage at cut and buried wells and the complexities of detecting and quantifying such leakage have been understudied. So far, studies on methane leakage at cut and buried abandoned gas wells did not account for the possibility that significant portions of leaking gas may be dispersed and oxidized in the vadose zone, which could have led to an underestimation of the actual occurrence and leakage rate. In the Netherlands, there are currently roughly 900 onshore oil and gas wells that have already been abandoned (Table 1) and the effectiveness of past and present abandonment procedures is largely unknown. Therefore, we selected 29 plugged, cut and buried wells for field investigation into the occurrence of gas leakage at these sites and to assess the importance of gas dispersion and oxidation in the vadose zone using static flux measurements above holes drilled into the surface, in addition to methane flux measurements and thorough concentration scanning at the surface. The aims of the study were to evaluate (1) the potential methane leakage from abandoned wells in the Netherlands and (2) the suitability of various methods for detecting and quantifying gas migration from abandoned wells that have been cut and buried to below the ground surface.

2. Materials and methods

2.1. Oil and gas wells in the Netherlands

The Netherlands has a long history of oil and gas production, and has been one of the major hydrocarbon producing countries in Europe. It owes this mainly to the discovery of two large reservoirs: the Schoonebeek oil field in 1943 and the Groningen gas field in 1959. In combination with the discovery of numerous smaller fields, this has led to the drilling of around 2500 hydrocarbon wells, of which >1700 onshore (Table 1). Gas production and drilling of new onshore wells in the Netherlands has slowed down in recent decades, because of depletion of reserves and induced seismicity in the Groningen area (van Thienen-Visser and Breunese, 2015). Currently, around 85% of all

Table 1
Breakdown of oil and gas well status in the Netherlands (NLOG, 2018).

	Active	Closed-in	Plugged/abandoned	Other/ unknown	Total
Onshore – oil	29	64	706	27	826
Offshore – oil	24	16	97	12	149
Onshore – gas	439	212	191	64	906
Offshore – gas	179	105	270	55	609
Total	671	397	1264	158	2490

onshore oil wells and 20% of onshore gas wells have already been abandoned (Table 1). The Dutch mining law regulates abandonment procedures of oil and gas wells. Besides prescribing detailed plugging procedures (Van Der Kuip et al., 2011), it also mandates that onshore wells are cut to at least three meter below the surface and buried (6 m below the seafloor for offshore wells). While this law does not prescribe any post-abandonment monitoring, operators still carry responsibility for the well according to Dutch civil law, should any deviation from the abandonment regulations be uncovered later (i.e., if leakage is detected) (Ministry of Economic Affairs, 2017).

Publicly available well integrity data from Dutch wells is limited. A 2011 assessment carried out by the State Supervision of Mines (SodM) uncovered well barrier failure in 1 out of 26 production wells and 3 out of 5 water injection wells for produced water (Vignes, 2011). However, well barrier failure does not necessarily equate to well integrity failure (King and King, 2013), given that there are often multiple barriers in place (e.g. several layers of casing and cement). Hence, the risk that these wells pose collectively for both groundwater quality and anthropogenic GHG emissions is unknown. Recently, ongoing gas leakage was detected at one borehole where a catastrophic blowout occurred while drilling this well in 1965, and as such it was not abandoned according to protocol (Schout et al., 2017). Thus far, this is the only known instance of methane leakage to groundwater in the Netherlands.

2.2. Well selection

Hydrocarbon well locations and other meta-data were obtained from the governmental 'Netherlands Oil and Gas Portal' (NLOG, 2018). Gas wells were prioritized over oil wells. No differentiation is made in the database between wells that have only been plugged and those that also have been cut and buried. Hence, a subset of such wells was identified by visual inspection of satellite imagery, at which point accessibility for fieldwork was also estimated. For example, locations that were built over already could not be included. Wells were selected from the various gas reservoirs spread throughout the Netherlands. Finally, as most abandoned well locations are situated on private land, accessibility depended on obtaining permission from the landowner during fieldwork. Ultimately, measurements were carried out at 29 locations (Fig. 1) during June and July of 2017: 28 gas wells and 1 oil well (RWK-14).

2.3. Surficial scanning

In the Netherlands, the locations of cut and buried hydrocarbon wells are not visually marked at the ground surface. Hence, they had to be located using GPS based on available coordinates. To decrease the chance of missing methane emissions from a leaking well, methane concentrations at the soil-atmosphere interface were first scanned in a circular area above the abandoned wells. A minimum radius of 15 m was chosen, which was estimated to be much larger than the uncertainty of the GPS and the given coordinates of the well. Scanning was performed using a Gazomat TDL-500 Inspectra Laser, which measures CH_4 concentrations real-time with a resolution of 0.1 ppmv and was checked for accuracy every day using a calibration gas cylinder (50 ppmv). To scan each site thoroughly and in a repeatable manner, the operator of the device was connected with a rope to a cylinder (1 m circumference) placed at the coordinates of the well and had to walk in circles around this cylinder while keeping the rope tight. The resulting spiraling pattern (known as the 'involute of a circle', Fig. 2) places the operator closer to the well every lap with a distance equal to the circumference of the cylinder.

A rod with a suction cup was attached to the inlet of the device (Fig. 2) that was held just above the soil surface during the scanning and moved from side to side while walking slowly. Additionally, the suction cup was placed directly on the soil at regular intervals and left for 5–10 s, after which methane concentrations and GPS coordinates were recorded. The locations of any deviating values were marked for subsequent analysis with static chamber flux measurements. The distance along the spiral in between measurements was decreased towards the well, such that the density of measurements was highest near the well location. Scanning in this manner yielded an overall average of around 160 data points per well. Maps of the recorded values for each well location are shown in Fig. S1.

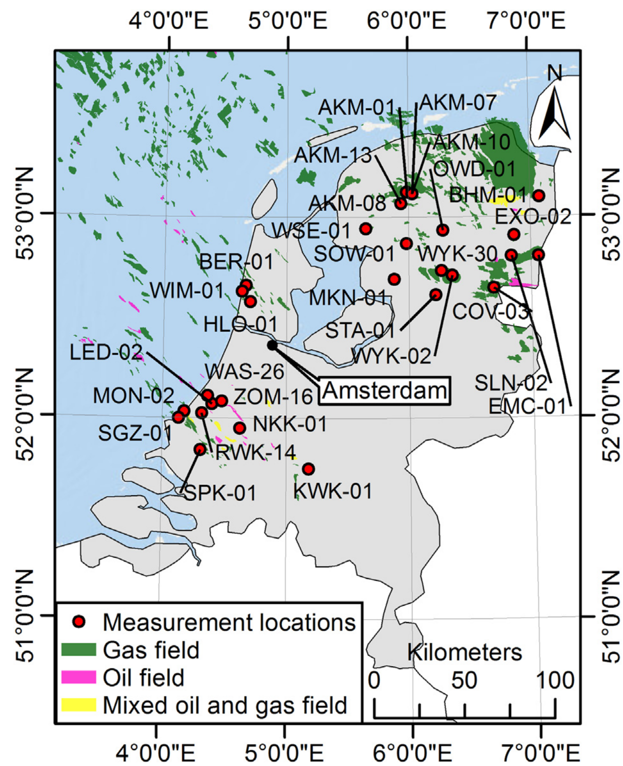

Fig. 1. Locations of the 29 investigated abandoned wells together with the oil and gas fields in the Netherlands (NLOG, 2018).

2.4. Static chamber measurements

Static chamber measurements were carried out directly above the buried remains of the well and, for 22 of the 29 well sites, a control

Fig. 2. Photo of the surficial scanning in action. Operator is attached to a wooden cylinder placed on the estimated location of the buried well, as marked with a red dot. The black line schematically shows the remaining path to be walked. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

location. For consistency, controls were taken at the starting point of the surficial scanning, i.e., a minimum of 15 m away from the well. This distance was expected to be larger than the uncertainty range associated with locating the well. However, the soil at this location would still be similarly affected by the abandonment procedure of the well (pad), local land use and soil conditions. Measurements were additionally conducted at locations that had elevated methane concentrations during the scanning.

A plexiglas cylindrical chamber (Fig. 3) with a height of 56 cm and a radius of 11.5 cm was used (volume of 23.25 L). To limit gas exchange with the atmosphere, the cylinder was placed on a plastic ring that was pushed 10 cm into the ground. Methane concentrations in the cylinder were recorded at regular intervals using the Gazomat TDL-500, also used for the surficial scanning. Both the inlet and outlet of this device were connected to the chamber with flexible tubing, such that measured gas was returned to the chamber (Fig. 3). The pumping rate of the device was 70 L hr^{-1}, which ensured mixing inside the cylinder. After completion of the measurements at the soil surface, 1 m deep open holes were drilled using a hand auger (5 cm diameter). Measurements were then directly repeated above the newly drilled holes (with the ring still in place).

The mass flow rate into the chamber was calculated using linear regression on the concentration versus time data (Eq. 1).

$$F = dC/dt{\cdot}V \tag{1}$$

where F is the CH_4 flow rate (mg hr^{-1}), dC/dt is the rate of change in CH_4 concentration in the chamber [mg cm^{-3} h^{-1}] and V the chamber volume [cm^3]. Similar to previous studies (Kang et al., 2016, 2014), only linear fits with an R^2 value >0.8 were considered. R^2 values below 0.8 only occurred when the concentration in the chamber remained close to the atmospheric methane concentration. Hence, the methane flow at these locations was assumed to be zero. Measurements were discontinued if no trend or change in concentrations inside the chamber was observed after 5 min. While the measurement resolution was 0.1 ppmv, experience learned that measured concentrations sometimes drifted slightly, going up and down within a bandwidth of around 0.5 ppmv. Assuming a temperature of 18 °C and a pressure of 1 atm, the minimum mass flow that would result in a positive determination within 5 min ($\Delta CH_4 \geq 0.5$ ppmv) was 0.09 mg hr^{-1}. In some cases the measurement period was extended to lower this detection limit slightly.

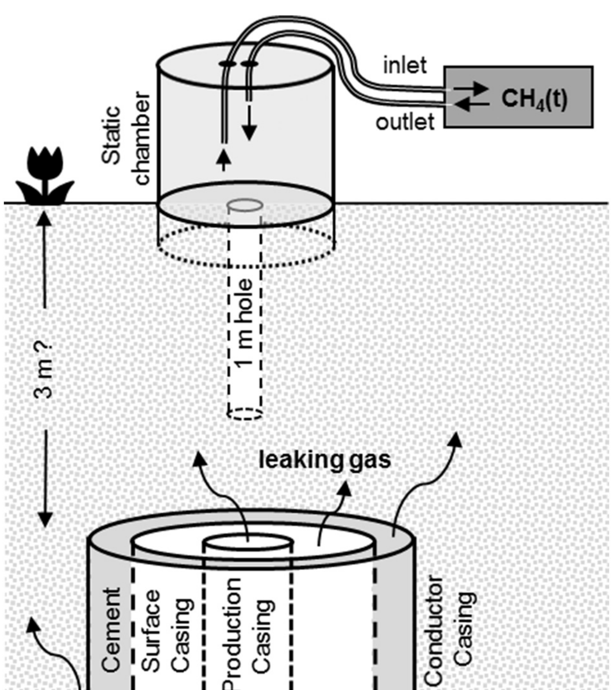

Fig. 3. Schematic representation of the static chamber measurements carried out above plugged, cut and buried wells. Chamber dimensions were 56 cm height, 11.5 cm radius, 23.25 L volume. Static chamber measurements were first carried before drilling the 1 m hole on which additional static chamber measurements were conducted. Figure not to scale.

2.5. Analysis of gas samples for stable methane isotopes

Gas samples for isotopic analyses were collected upon completion of the static chamber measurements by placing 1 L tedlar bags in a sealed box attached directly to the static chamber. Vacuum was created in this box using a foot pump, causing the sample bags to fill with gas from the chamber. As measured fluxes were generally higher at 1 m depth, it was decided to collect samples after these measurements rather than those at the surface. Samples were collected at additional locations if methane leakage from the abandoned well was suspected. Analyses of the samples were carried out at the Institute for Atmospheric and Marine Research (IMAU) of the Utrecht University. Methane was extracted as described in Röckmann et al. (2016): methane carbon ($\delta^{13}C$-CH_4) and hydrogen (δ^2H-CH_4) stable isotope ratios were determined in separate measurements by Isotope Ratio Mass Spectrometry (IRMS) on a Thermo Finnigan Delta plus XP. The sample mixing ratio and isotopic composition were determined by comparing the measurements to that of a known reference air that was extracted in the same way.

2.6. Detailed characterization of methane leakage at the MON-02 site

Thermogenic methane was observed at well MON-02 following the previously described procedures. This site was therefore characterized in more detail. Well MON-02 produced from the 'Monster' gas field (Fig. 1), a small gas accumulation in the West Netherlands Basin, and was abandoned in the 1990s. The reservoir consists of a series of Triassic layers known together as the Main Bundsandstein Subgroup, and is situated at a depth around 2800 to 3000 m. Starting at the coordinates of the abandoned wellbore, static chamber measurements were carried out in each cardinal direction with a 2 m spacing. Once a negligible flux was identified at 1 m depth, measurements were continued in the next direction. This procedure led to 14 measurement locations that form a grid like pattern, with each measurement representing an area of 2 by 2 m. In addition to the procedure described in Section 2.4, static chamber measurements were also conducted at 2 m depth, slightly above the groundwater table at this location (2.2 mbgl). This was done by extending the already drilled holes to 2 m once the 1 m deep measurements were completed. Furthermore, PVC tubes with the same length and diameter (5 cm) as the drilled holes were placed inside the holes and connected directly to the chamber so that flow into the chamber could only occur from the bottom of the tube. A sandy soil profile was encountered throughout the site. Samples for isotopic analysis were collected at the end of seven measurements of a single transect: three at 1 m depth and four at 2 m depth.

In total, 28 static flux measurements were carried out (Table S.2). Regression was performed over a linear part of the data ($R^2 \geq 0.8$) of at minimum 50 s in order to calculate the flow rates. To estimate the total leakage flux from the well the methane flow rates were converted to a measured mass flux (Eq. 2):

$$J_m = F/A_t \tag{2}$$

where J_m is the measured CH_4 flux [mg cm^{-2} h^{-1}] and A_t is the surficial area of the opening of the tube [cm^2]. Due to the lack of a porous medium inside the tube, the diffusive flux towards the chamber was enhanced compared to the initial, undisturbed soil condition. To account for this, measurements were scaled by the ratio of the diffusion coefficient of methane in sandy soil to that in air, yielding an approximation of the undisturbed flux:

$$J_u = (D^e/D^a)\, J_m \tag{3}$$

where J_u is the estimated undisturbed methane flux [mg cm^{-2} h^{-1}] and D^e and D^a the effective diffusion coefficient in sandy soil and the diffusion coefficient in air, respectively [cm^2 h^{-1}]. A D^a of 726 cm^2 h^{-1} was assumed according to the method of Fuller et al. (1966) for CH_4 at 18 °C and 1015 mbar. The effective diffusion coefficient in sandy soil was calculated as follows (e.g. Scanlon et al., 2002):

$$D^{eff} = \phi\, \tau\, S_g\, D_a \tag{4}$$

where ϕ is the porosity [−], S^g is the saturation of the gas phase [−] and τ the tortuosity factor (determined according to Penman, 1940a, 1940b. A porosity of 0.3 and soil gas saturation of 0.9 were assumed. Finally, the total leakage flux from the well was calculated by means of a nearest neighbor interpolation, at 1 and 2 m depth separately: first, the estimated undisturbed flux for each measurement location was extrapolated to the 2 × 2 m square area surrounding it and then all data points were summed.

3. Results

3.1. Surficial scanning

Surficial scanning of methane concentrations at the soil-atmosphere interface was carried out fully at 19 and partly at 5 of the 29 selected well sites (Fig. S1). At 10 locations obstructions such as canals or dense undergrowth either partially or fully limited the required mobility (see for example Fig. 4B). During the course of most measurements, observed concentrations tended to drift around the atmospheric methane concentration (~1.8 ppmv) between roughly 1.1 and 2.5 ppmv. Thus, observed patterns of both increasing and decreasing concentrations could not be related to locally enhanced methane emissions or uptake by the soil as long as the concentration changes remained within this bandwidth (e.g. Fig. 4A and Fig. 4B). Isolated locations with concentrations >2.5 ppmv were observed at 6 well sites (Fig. S1). In most cases, these observations were temporary concentration spikes and could not be reproduced on subsequent measurements. However, these locations were marked for later investigation using the static chamber setup. Only while scanning well site NKK-01 elevated methane concentrations up to several hundred ppmv were observed that also sustained over a longer time period (Fig. 4C).

3.2. Static chamber measurements at surface and 1 m depth

The static chamber measurements carried out at the soil surface directly above the 29 buried wells revealed only a single location with a positive methane flow rate: AKM-07 (Fig. 5, Table S.1). In contrast, flow rates were negative at the surface above four wells, and no measurable methane flow was recorded above the remaining wells. The surficial static chamber measurements also revealed two positive and one negative flow rate at the control locations; flow could not be detected at the remaining control locations. Negative flow rates are interpreted as methane uptake from the soil caused by methane oxidation in the vadose zone. At 1 m depth, positive methane flow rates were observed more frequently than at the surface while negative flow rates were only observed at two control locations. Overall, 15 of 29 well locations and 10 of 22 control locations had a positive methane flow rate at 1 m depth (Fig. 5). With the exception of well NKK-01, measurements carried out at locations that were marked because of concentration outliers observed during the surficial scanning did not yield appreciably higher flow rates. Hence, these measurements were not considered for further interpretation.

Methane flow rates at 1 m depth were larger above the buried wells than at their respective control for 6 locations. Conversely, control measurements were larger at 8 other locations (Fig. 5). The remaining 8 locations where control measurements were conducted had undetectable flow rates both above the buried wells and at the control locations. As can be seen in Fig. 5, the variability in flow rate between the well and control locations was such that the difference between them could not be relied upon to attribute measured flow rates above the buried wells to well integrity failure. A clear positive outlier was, however, observed at well MON-02 with a flow rate of 1418 mg hr^{-1} at 1 m depth. This is more than two orders of magnitude higher than the next highest flow rate measurement in the entire study. Furthermore, this high flow rate contrasted with the negative flow rate measured at 1 m depth at the control location, the undetectable flow rates at the surface and the lack of elevated concentrations recorded during the surficial scanning (Fig. 4A).

3.3. Analysis methane stable isotopes

Only the sample collected after the flux measurement at 1 m depth above well MON-02, where the highest single flux measurement was observed, had a clear thermogenic origin (δ^{13}C-CH4 = −26.6‰ and δD-CH4 = −149.7‰, Fig. 6). This is convincing evidence that leakage of natural gas caused by well integrity failure was occurring. Samples from other locations with high flow rates had isotopic compositions

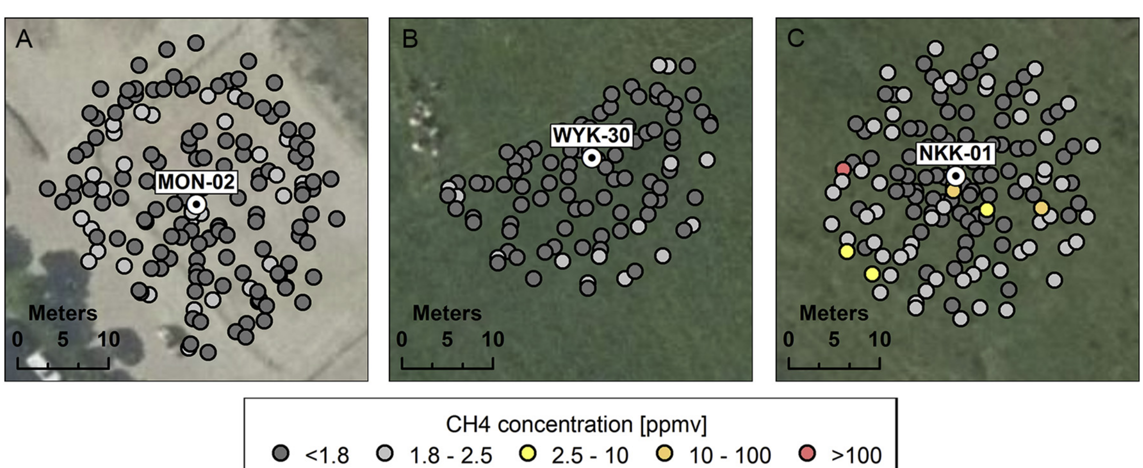

Fig. 4. Three examples of the results obtained during surficial scanning of methane concentrations: A) MON-02, B) WYK-30, where only half the scanning spiral could be executed and C) NKK-01, where several concentration outliers were identified that were marked for static chamber flux measurements.

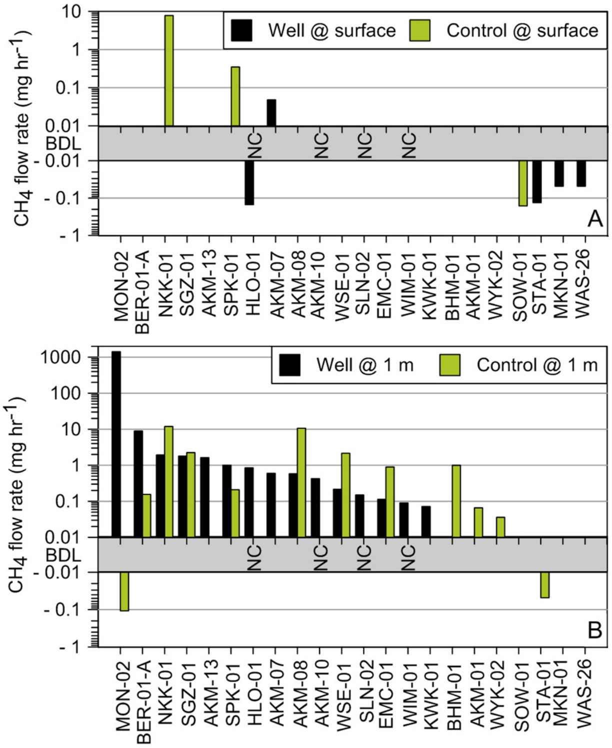

Fig. 5. Methane flow rates at the surface (A) and at 1 m depth (B) as determined by static chamber flux measurements. Locations are shown that have at minimum one non-zero measurement (22 out of 29 locations). Grey area is below the detection limit ('BDL'), locations where no control measurements were carried out are labelled with ´NC´.

indicative of a biogenic origin, with a $\delta^{13}C-CH_4 < 50‰$ and a $\delta D-CH_4 < -225‰$, regardless of whether the fluxes above the well or at the control location had given higher fluxes. A biogenic gas source was also confirmed at well NKK-01, the only location where high methane concentrations where recorded during the surficial scanning that also resulted in positive flow rate measurements at the surface. The bacterial methane is likely related to the presence of organic-rich formations, abundant in the shallow subsurface of the Netherlands. Many oil and gas fields in the Netherlands are found in the coastal provinces, where Holocene peat and clay deposits are also present (Wong et al., 2007). Methane emissions from these deposits are well known (Van Den Pol-Van Dasselaar et al., 1999). Samples from locations with intermediate flow rates showed isotopic compositions that can be explained by mixing of biogenic and atmospheric methane, while sites with zero or low flow rates showed an atmospheric origin with $\delta^{13}C-CH_4$ and $\delta D-CH_4$ values of around −47‰ and −93‰, respectively (Fig. 6).

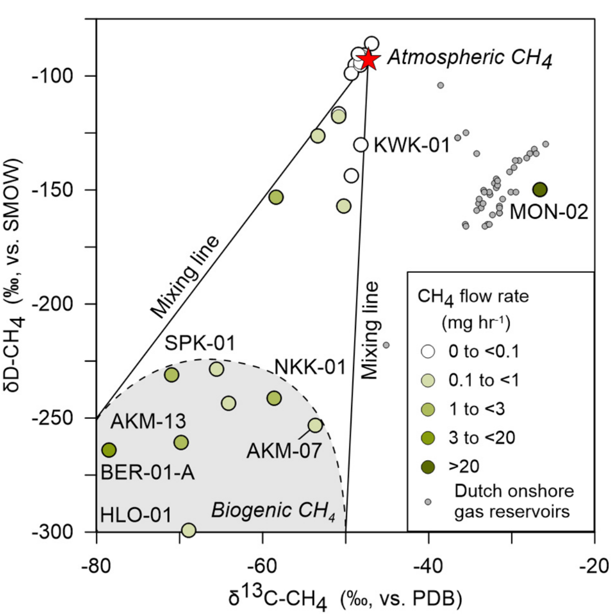

Fig. 6. Carbon and hydrogen isotopic composition of methane in samples collected after completion of static chamber measurements at 1 m depth. Grey dots show isotopic composition of samples taken from onshore Dutch gas reservoirs (NLOG, 2018). Isotopic composition of northern-hemisphere atmospheric methane based on Rice et al. (2016).

3.4. Detailed characterization of methane leakage at the MON-02 site

The detailed characterization carried out at well MON-02 showed that the estimated undisturbed methane fluxes were significantly higher at 2 m depth than at 1 m depth (Fig. 7). At the location directly overlying the coordinates of the abandoned wellbore (A1), the estimated undisturbed flux at 1 m depth was 9.4 g m^{-2} h^{-1} compared to 19.6 g m^{-2} h^{-1} at 2 m depth. While the maximum flux at 1 m depth was located at the coordinates of the well, the maximum flux at 2 m depth was located almost 3 m to the south (B2) and was 4.5 times larger than the maximum flux at 1 m depth: 40.4 g CH_4 m^{-2} hr^{-1}. Considerable lateral spreading is observed with fluxes >2 g m^{-2} hr^{-1} measured up to 4.5 m distance from the well coordinates at 1 m depth and up to 6 m at 2 m depth. If subsurface methane concentrations had solely been the result of diffusion from a buried point source through a homogeneous, sandy porous medium (as encountered at the site), spreading would have occurred in semi-spherical manner in all directions equally. However, this is not the case, as illustrated by the fact that the fluxes just 2 m northeast of the coordinates of the well (O1) were zero at both 1 and 2 m depth. Nearest neighbor interpolation of the data yielded a total leakage flux estimate of 111 g hr^{-1} at 1 m depth and 443 g hr^{-1} at 2 m depth. At atmospheric pressure and a temperature of 18 °C, this is equal to a volumetric flux of 4.0 and 15.8 m^3 day^{-1}, respectively. Furthermore, it suggests that a reduction in flux of 74% had occurred from 2 to 1 m depth.

Isotopic compositions of the 7 collected samples at MON-02 varied significantly (Fig. 8). The isotopic composition of methane from the Monster gas reservoir is known from a publicly available analysis of a gas sample collected from nearby gas well MON-03 (NLOG, 2018). The sample collected at the location with the lowest flux (A4-2 m) lies close to the mixing line of gas from the reservoir and atmospheric methane. The samples that are isotopically most similar to that of the reservoir are three samples collected at 2 m depth at the locations with the highest flux measurements (A1-2 m, A2-2 m and A3-2 m). Relative to these three samples, two of the samples from 1 m depth (A1-1 m and A3-1 m) are enriched in both their carbon and hydrogen isotopic composition. This is indicative of the occurrence of aerobic microbial methane oxidation as the isotopic fractionation factors observed for aerobic methane oxidation in Dutch and German soils ($\alpha^{13}C = 1.008$ and $\alpha D = 1.0039$, Bergamaschi et al., 1998) qualitatively match the shift from the samples collected at 2 m depth to those at 1 m depth (Fig. 8). Following the methods described in Whiticar (1999), the observed shift can be related to a residual methane fraction of 75%. In other words, methane oxidation can account for up to 25% of the reduction in methane flux as observed from 2 to 1 m depth.

The isotopic composition of sample A2-1 m, which is isotopically depleted compared to the reservoir gas, represents an outlier that cannot be explained by leakage of gas from the Monster gas reservoir. Possibly the leaking gas here does not originate from the reservoir, but rather from an intermediate formation. The most likely candidate is the Holland Greensand Member, a known gas-bearing formation in the Netherlands located here at a depth of 1420 to 1481 m. The isotopic composition of this formation is known from two samples taken from a nearby small gas field (Fig. 8) and matches that of the sample taken at A2-1 m relatively well.

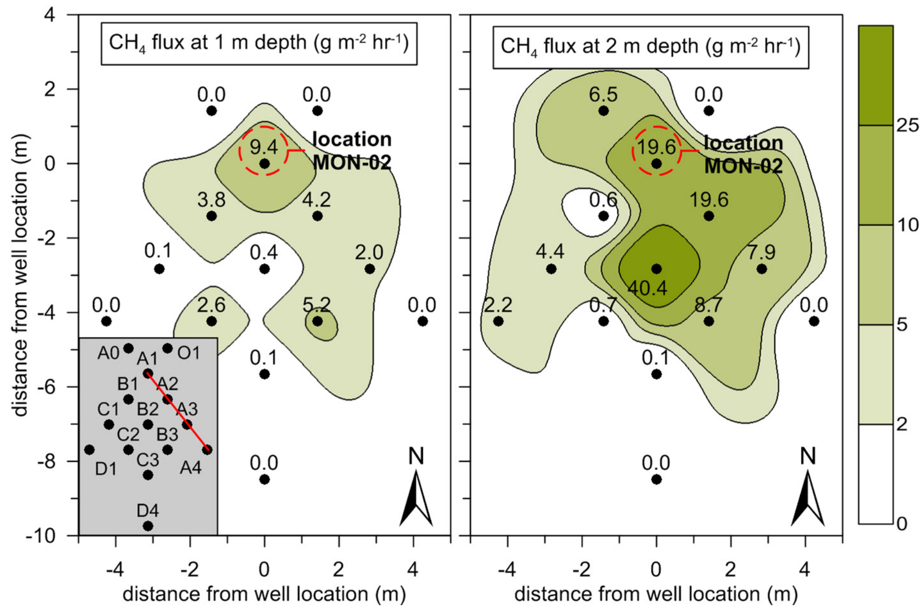

Fig. 7. Results of the detailed investigation at MON-02: values signify the estimated undisturbed fluxes at 1 m depth (left) and 2 m depth (right), contours depict the underground spreading of leaked methane but are speculative. Grey inlay shows the label for each position, the red line indicates the locations at which samples were collected for isotopic analysis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Discussion

4.1. Detecting gas leakage from cut and buried abandoned wells

As shown by the results of our study, the occurrence of gas leakage at cut and buried abandoned wells is more easily missed when relying on surface measurements only, as compared with abandoned wells that still have existing wellhead infrastructure protruding to the surface. Besides additional uncertainties on the exact location of the buried well, particularly in the absence of surface markers (as is the case in the Netherlands), leaking methane may become dispersed and oxidized as it migrates from the leakage point towards the surface, not necessarily at the well coordinates due to preferential flow paths. When a leak originates below the groundwater table, upwardly migrating methane can be additionally attenuated by dissolution into groundwater (Cahill et al., 2017). Obviously, when gas leakages result from well integrity failure at greater depth and the gas migrates along the outside of the casing these processes would equally affect methane transport to surface (Dusseault and Jackson, 2014), regardless of whether a well is abandoned, cut and buried, or still protruding to the surface. A combination of these processes caused leakage from a gas well in Colorado, USA, to be undetectable by surficial flux measurements and soil gas probes installed to 60 cm depth, while methane concentrations in the underlying groundwater were above air-saturation levels (McMahon et al., 2018). Subsurface (vadose zone) measurements using dedicated soil gas wells have previously been employed in soils surrounding oil and gas wells that were not cut and buried by (Lyman et al., 2017). However, while this allows for continued and precise monitoring of soil gas concentrations, our method is less costly and time consuming and therefore enables relatively quick screening at multiple locations.

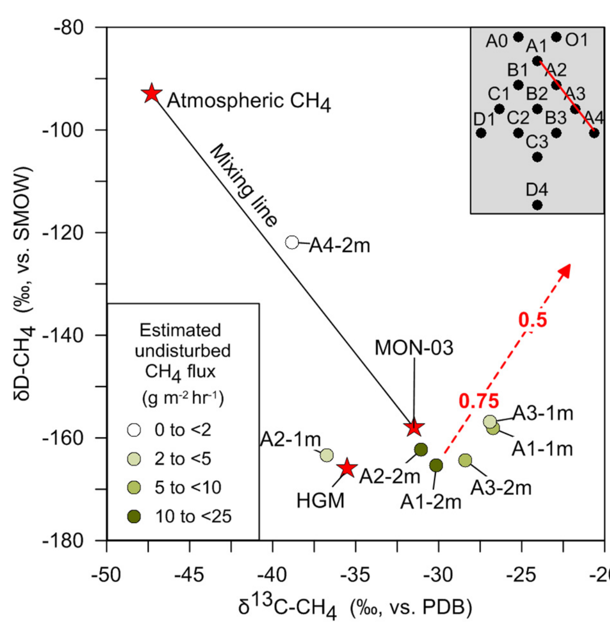

Fig. 8. Isotopic composition and estimated undisturbed flux of seven samples collected as part of the detailed investigation at MON-02 (sampling positions indicated by red line in figure inlay). Two thermogenic end members are shown: Monster reservoir gas (MON-03) and gas from the intermediate Holland Greensand Member (HGM; NLOG, 2018). Red dashed line shows the estimated effect of aerobic methane oxidation, red labels the estimated residual methane fraction. Calculation based on isotopic fractionation factors found in soils above Dutch landfills ($\Delta D/\Delta C = 4.8$, Bergamaschi et al., 1998). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Methane leakage from a cut and buried wellbore may also be masked by mixing with naturally occurring biogenic methane. Such emissions can be highly spatially variable as a result of soil heterogeneities and preferential pathways (Hendriks et al., 2010). This is illustrated in our study by the detection of hotspots of elevated methane concentrations during the surficial scanning at several locations above one well (NKK-02; Fig. 4C), but where isotopic analysis revealed a shallow biogenic origin. Furthermore, local variabilities in biogenic methane fluxes also explain the lack of correlation between the magnitude of the fluxes measured at surface and 1 m depth as well as their controls (Fig. 5). Given these conditions, isotopic confirmation of methane origin is critically important in preventing false negatives or positives in the detection of subsurface gas leakage from a well. At the one well in this study for which gas leakage was confirmed, both the surficial scanning and the surface static chamber measurements did not yield any noticeable indication of leakage (MON-02; Fig. 4A, Fig. 5). This emphasizes the importance of subsurface measurements of methane flow rates or concentrations, above the level from where leaks could be occurring, as dispersion and oxidation will have had less impact on the methane flux than at the surface. Also, for conditions of high background biogenic methane production, the dilution and mixing of the isotopic signature of the leaking gas would be smaller.

Although oxidation and dispersion decrease the detectability of methane leakage, isotopic analysis can, in addition to determination of origin, aid in determining the relative importance of these two processes in the total attenuation of the subsurface methane flux from a cut and buried well. As shown by the enrichment of the methane isotopic composition (Fig. 8) concurrent with the decrease in flux between 2 and 1 m below ground surface for the gas leakage occurring at well MON-02 (Fig. 7), aerobic methane oxidation accounted for roughly 25% of this decrease. Hence, around 75% of the decrease in methane flux towards the surface was attributable to dispersion for that well site. Negative fluxes, resulting from the net oxidation of methane were indeed observed at several of the investigated locations, including at the control location of well MON-02 (Fig. 5). Furthermore, the widespread oxidation of subsurface methane fluxes (whether biogenic or thermogenic) is also suggested by the consistently larger fluxes measured at 1 m depth than those measured at the surface for the other locations investigated (Fig. 5). The extent to which dispersion and oxidation affect the methane flux from subsurface well leakage will however strongly vary between different well locations, depending on the depth of the leakage point, as well as a range of local physical (e.g. water content) and biochemical (e.g. redox conditions, microbial population) conditions and the heterogeneities therein.

4.2. Estimation of the total methane leakage flux from cut and buried abandoned wells

At abandoned wells that are not cut and buried, sealing the wellhead off from the atmosphere allows for accurate measurements of the total leakage flux (assuming there is no gas migration outside the casing). At cut and buried wells however, quantification of the total flux is more complex as each measured flux represents an unknown portion of the total flux. Furthermore, while necessary for the detailed characterization of leakage at MON-02, carrying out flux chamber measurements from holes drilled into the soil introduces additional uncertainty by disturbing soil properties and the initial methane concentration distribution in the vadose zone. The three most notable sources of uncertainty include first that we had to compensate for the lack of a porous medium in the tube installed into the drilled holes by scaling the measured flux with the ratio of the effective diffusion coefficient of methane in soil over the diffusion coefficient of methane in pure air. This requires knowledge of the porosity, tortuosity and gas saturation of the porous medium, which we did not determine in the field and hence had to be approximated. Second, the measurements for the detailed investigation did not yield strictly linearly increasing concentration versus time data. Hence, regression analysis had to be carried out on the linear part of the data for that particular flux measurement. The non-linear behavior was likely caused by a combination of factors, including the alteration of soil properties due the drilling, non-vertical and possibly some advective inflow of methane into the tubes, and disruption of the initial methane concentration profile in the soil. Third, interpolation of the measurements at 2 m depth assumes that methane fluxes are strictly vertical whereas in reality there is an important lateral component (as also shown by the results of our study). Further, the interpolation assumes that the flux is more or less spatially distributed and not dominated by fluxes through preferential flow paths. We believe this to be generally valid here given the sandy conditions and the lack of concentration outliers observed during the surficial scanning.

Given the number of sources of uncertainty and the assumptions required under these non-ideal subsurface leakage conditions, our estimation of the flux at 2 m depth (443 g CH_4 hr^{-1} or 15.8 m^3 CH_4 d^{-1}) should be considered an indicative value only. Also, vadose zone processes have been shown to lead to considerable temporal variation of methane fluxes measured by static flux chambers (Forde et al., 2018), and our measurements thus only represent a snapshot of the total leakage flux at one moment in time. Longer term monitoring would be recommended to better constrain the average flux from a leaking well. Future modelling and/or experimental studies may help to determine the validity of the approach taken for the estimation of total leakage flux. In comparison to leakage flux estimates in other studies, our total flux estimate at two-meter depth is larger than any of the fluxes identified at plugged wells by both Townsend-Small et al. (2016) and Kang et al. (2016). However, the flux estimate is in the same order of magnitude as the maximum fluxes they observed at unplugged wells. In comparison, surface casing vent flow fluxes appear to be much larger on average: fluxes larger than 10 m^3 day^{-1} were observed at 32% of 11,394 conventional wells with reported leakage issues in Alberta and British Columbia, Canada (Nowamooz et al., 2015). Therefore, our indicative total flux estimate does not seem unrealistic.

4.3. Observed well leakage frequency in context

This study revealed that methane leakage caused by integrity failure had occurred at 1 out of 28 cut and buried abandoned gas wells (3.6%). While this value is at the low end of the large range reported for well integrity failure in the review by Davies et al. (2014), it is considerably higher than the 0.06–0.15% found by Sherwood et al. (2016) in Colorado, USA. However, their findings were based on water chemistry analyses, which is a more indirect measurement and may therefore have contributed to a relatively low leakage frequency. The observed well leakage frequency is larger than that found by Townsend-Small et al., 2016 (0.8%) for plugged and abandoned oil and gas wells, part of which had also cut and buried, but lower than both the 30% reported by Boothroyd et al. (2016) for plugged, cut and buried wells in the UK and the 62% reported by Kang et al., 2016 for plugged wells that had not been cut and buried. These differences are likely caused by a combination of factors such as the local geology, well type, well construction, well abandonment procedures (King and King, 2013) and the type of monitoring method employed. Here, the amount of wells studies likely plays a role, too. More studies are needed in order to obtain proper insight in the statistics of well integrity failure for the Netherlands as well as globally.

4.4. Environmental impact of abandoned gas wells in the Netherlands

Given the limited amount of wells studied, extrapolation of the results can only be done tentatively. Using binomial statistics, the lower and upper probability that could have still resulted in 1 positive detection within one standard deviation were calculated to convert our findings to a range. This yields a well integrity failure rate for cut and buried gas wells in the Netherlands of between 1.4% to 9.0%, which is equal to a range of between 3 and 17 leaking wells when extrapolating to all currently abandoned gas wells ($n = 191$) and between 12 and 81 leaking wells when extrapolating to the total amount of onshore gas wells ($n = 906$). Assuming each well leaks with the estimated flux at 2 m depth at well MON-02, this gives a rough national estimate of between 10 and 66 ton CH_4 yr^{-1} emitted for the abandoned gas wells, or between 48 and 315 ton CH_4 yr^{-1} for all gas wells. This is equal to between 0.14 and 0.95% of the total amount of CH_4 estimated to have been emitted by the Dutch energy sector in 2015, and 0.005–0.035% of the total anthropogenic CH_4 emissions in the country (Environmental Data Compendium, 2017). In other terms, the maximum estimated value is equal to the methane produced by roughly 500 Dutch milk cows (~133 kg CH_4 yr^{-1} cow^{-1}; Velthof et al., 2016). Concluding, it is improbable that the methane leaking from these wells contributes significantly to current anthropogenic GHG gas emissions in the Netherlands.

In the case of cut and buried wells, part of the leaking methane is converted to CO_2 as a result of aerobic oxidation. Hence, combining methane concentration measurements with CO_2 measurements would be required to fully assess GHG emissions. Given that methane has a global warming potential 24 times that of CO_2 (Myhre et al., 2013), CO_2 emitted in this manner is likely insignificant for our study area. Notably, leakage from cut and buried oil and gas wells does have to be considered in the context of explosion hazards. This is illustrated by the MON-02 case reported in this study, where the intended construction of several new houses was upheld to repair the leaking abandoned well and ensure the safety of the new buildings.

5. Conclusions

Methane leakage from cut and buried abandoned oil and gas wells is an understudied environmental hazard, and the detection and quantification of leakage at such wells is complex. To assess environmental impact and effective monitoring methods, 28 cut and buried gas wells and 1 oil well were investigated in the Netherlands. Leakage was observed at 1 gas well (MON-02), constituting the first observation of well integrity failure at a properly decommissioned gas well in the country. Neither the surficial methane concentration scanning nor the static flux measurements carried out at the surface proved capable of detecting the leaking gas. However, static flux measurements above 1 m deep holes drilled into the soil were an effective method. In general, measured flow rates were consistently larger at depth than at the surface. Control measurements showed that naturally occurring biogenic methane fluxes were spatially highly variable. Therefore, the origin of measured fluxes could not be determined based on flux magnitude alone, and analyses of methane stable isotopes were required to distinguish between a biogenic or thermogenic origin.

A detailed characterization at the leaking well, consisting of static chamber measurements in conducted in a 2×2 grid from holes drilled to 1 and 2 m depth, showed that measured flow rates diminished rapidly towards the surface. Enrichment of the isotopic composition concurrent with this decrease confirmed that besides dispersion, aerobic methane oxidation accounted for roughly 25% of this decrease. Converting the flow rate measurements in the vadose zone to a total flux estimate is not trivial, as each measurement represents only a small portion of the total flux and the drilling of the holes required for the measurements disturbs the soil properties. Nevertheless, an indicative value was computed by converting each measurement at 2 m depth to estimates of the 'undisturbed' flux and then interpolating using nearest neighbor interpolation. This approach yields a value of 443 g CH_4 hr^{-1} emitted from the well. Extrapolating this number to all - abandoned or not - gas wells ($n = 906$) in the Netherlands and taking an uncertainty range of 1.4 to 9.0% for the well integrity failure rate yields a first estimate of 48–315 ton CH_4 yr^{-1}. This is negligible compared to the yearly methane emissions from the energy sector (0.14–0.95%) and overall anthropogenic methane emissions (0.005–0.035%) in the Netherlands.

Taken together, these findings support the need for subsurface measurements when trying to detect and quantify methane leakage from cut and buried gas wells, and have important implications for past and future studies aimed at detecting methane leakage at such wells. However, more research is required to assess whether point measurements of vadose zone methane fluxes can be reliable converted to estimates of the total leakage flux from a cut and buried well. In spite of considerable uncertainty, our results show that leakages from gas wells are unlikely to contribute significantly to GHG emissions in the Netherlands. However, the potential explosion hazard caused by methane leakage from cut and buried abandoned wells needs to be taken into account when planning construction projects.

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