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The Study of Gas Emission on Natural Gas Leak Localization

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Abstract - The aim of this study is to measure gas emission (CH_4 and CO_2) coming from natural gas leak localization in the soil. Natural gas is injected into soil in different depths and then analyzed by gas detector and micro gas chromatography to know the values CH_4, C_2H_6, C_3H_8, C_4H_{10}, C_5H_{12}, O_2, N_2 and CO_2 which is spread out into soil. When there are leaks in the soil, methane (CH_4) will spread out underground. Methanotropic bacteria will use this natural gas as an energy source and transform it into carbon dioxide. The micro gas chromatography data was found that the pipes injected from 20 cm leak are 77.16% CH_4 loss in 70cm depth, 73.15% in 50cm depth and 14.08% in 20cm depth. And the pipes injected from 30 cm leak are 20.27% in 30 cm depth and 65.13% in 60 cm depth. Then, the pipes injected from 50 cm leak are 23.40% in 30 cm depth and 47.40% in 60 cm depth. The leak source is in 80 cm depth.

Key words : methane; carbon dioxide; ampoule; gas detector; micro gas chromatography

INTRODUCTION

Methane is an important greenhouse gas thought to contribute to the global warming. On a global scale the contribution amounts to roughly 2 percent of the total equivalent emission rate of all greenhouse gases.

There are varieties of methods that can detect methane pipe line leaks, ranging from manual inspection using trained dogs to advanced satellite based hyper spectral imaging (9 & 21). The various methods can be classified into non-optical and optical methods. The primary non-optical methods include acoustic monitoring (26 & 43), gas sampling (44), soil monitoring (47), flow monitoring (7 and 47) and software based dynamic modelling (13 &17).

The acoustic monitoring techniques, which is utilizing acoustic emission sensors to detect leaks based on changes in the background noise pattern (47). This method is easy to use in various sizes of pipes. But, it requires a large number of acoustic sensors to monitor an extended range of pipelines. Unfortunately, this technology is unable to detect small leaks that do not produce acoustic emissions at level substantially higher than the background noise. Furthermore it may be difficult to distinguish a leak from a service line noise emission. Thus this method is adapted mainly for pipes without customers.

Soil monitoring method is a specific technique where pipeline is firstly inoculated with a small amount of tracer chemical. This tracer chemical will seep out of the pipe in the event of a leak. This is detected by dragging instruments along the surface above the pipeline. This method has high sensitivity because it includes a very low false alarm, but on the contrary, it is costly.

Flow monitoring devices measure the rate of change of pressure or the mass flow at different sections of the pipeline. Where there is a different value of pressure or mass flow, it could detect a potential leak. This method is low cost, but it cannot detect the leak location and give high rate of false alarms.

Software based dynamic modelling monitors various flow parameters at different locations along the pipeline. These flow parameters are then included in a model to determine the presence of natural gas leaks in the pipeline. The advantage is it has the ability to check and monitor continuously where the leaks location. However, it has a high failure in giving alarms and is costly for monitoring a large network of pipelines (47). This method, which is an improvement of the flow monitoring method, may be efficient on transmission lines but not on distribution.

The most frequently used methods by distribution system operators are gas-sampling methods. Gas sampling methods typically use a flame ionization detector housed in a hand held or vehicle mounted

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probe to detect methane or ethane. The primary advantage of flame ionisation detectors is that they are very sensitive to very small concentrations of gasses. This method can detect a very slow and limited gas leaks to the local area from which the gas is drawn into the probe for analysis. Therefore, the cost is expensive for monitoring long pipelines.

The leak environment is very important especially regarding to the possibility for the leaking gas to ingress in a building. This would be possible, and then the leak is considered as very dangerous.

The specific gravity of natural gas is one kind of characteristics to control gas leak underground, it is approximately 0.6 which is, therefore, lighter than air. This property facilitates the venting and dissipation of natural gas leakage into the atmosphere. The flammable range of natural gas is approximately 5 – 15% gas in air.

TEORITICAL STUDY

1.1 The way to detect Leakage

1. Qualification personnel
   Gas leakage surveys should be performed by personnel who are qualified by training and experience in the type of survey being performed. They should be familiar with the characteristics of natural gas in the system and trained in the use of leakage detection instruments.
2. Report from outside sources
   Any notification from an outside source (such as police or fire department, other utility, contractor, customer or general public) reporting an odor, leak, explosion or fire which may involve gas pipelines or other gas facilities should be investigated promptly. If the investigation reveal a leak, Then the leak is automatically considered as Class 1 as it as been reported by third chapter. It is repaired at once.
3. Odors or indications from foreign sources
   When leak indications are found to originate from a foreign or facility or customer own piping, prompt actions should be taken where necessary to protect life and property. Potentially hazardous leaks should be reported promptly to the operator of the facility and, where appropriate, to the police department, fire department or other governmental agency. When the company's pipeline is connected to a foreign facility (such as the customer's piping), necessary action (such as disconnecting or shutting off the flow of gas to the facility) should be taken to eliminate the potential hazard.
4. Leakage surveys and test methods
   The following gas leakage surveys and test methods may be employed, as applicable, in accordance with written procedures.
   - Subsurface gas detector survey (including bar hole surveys)
   - Bubble leakage test
   - Pressure drop test
   - Ultrasonic leakage test

Natural gas leaks are a hazard above a certain concentration in the air namely LEL (Lower Explosive Limit). The mixture can burn and therefore explode. Between the LEL and the UEL (Upper Explosive Limit) i.e. 5 – 15% the air is still breathable (35).

1.2 Methanotropic Bacteria

Methanotrophs are a unique group of methylotrophic bacteria which utilize methane as their sole carbon and energy source (12, 29). They use enzyme *methane mono oxygenase (MMO)* (33). This MMO can cometabolize or transform non-growth substrates by either growing or resting cells. Cometabolism is a result of non-specific MMO activity towards organic compounds that do not serve as carbon or energy sources. These organisms have been isolated from a wide variety of environments including soils (40), sediments (39), landfills, ground water (15), seawater (19, 29, and 37), peat bogs (12, 28), hotsprings (3,4), plant rhizosphere (16), salt reservoirs (24) and the Antarctic (8) and Bioremediation (33).

MMO is the enzyme responsible for the oxygenative catalysis of methane to methanol (11).

The methanotrophs can cometabolize many aliphatic compounds, alkanes and aromatic compounds. (33)

Methanotrophs were initially grouped according to their morphology, type of resting stage, intracytoplasmic membrane structure and physiological characteristics (40).

Mostly, methanotrophs gain their energy for growth from the oxidation of CH_4 to CO_2 by consuming CH_4 as the energy source. Methanotrophs plays an important role in oxidizing methane in natural environment.

The overall reaction of methane oxidation is:

$CH_4 + 2\,O_2 \rightarrow CO_2 + 2H_2O$ (ΔG^o = -778 kJ/mol CH_4) (41)

According to the reaction above, it seems that methanotrophic bacteria require both single carbon compounds and oxygen to survive, combining the two to form formaldehyde, which is then incorporated into organic compounds. They also characteristically have a system of internal membranes within which methane oxidation occurs.

Figure 1 below shows the pathway for methane oxidation

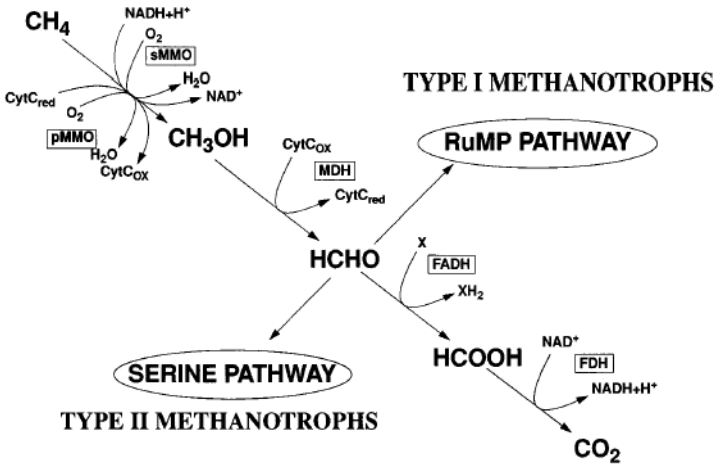

Figure 1. Pathway for the oxidation of methane and assimilation of formaldehyde (33)

Methanotrophs occur mostly in soils, and are especially common near environments where methane is produced. Their habitats range from oceans, mud, marshes, underground environments, soils, rice paddies, etc. (41)

The activity of methanotrophs bacteria depends on the presence of methane and oxygen concentration and also they tend to be limited in thin soil horizons, typically in the top 30 cm of soil. It means that they are limited in distributing the downward diffusion of atmospheric oxygen and the upward diffusion of methane (5, 6, 4, 23 and 27). This is very important as gas mains are usually below that depth, typically in 80 cm depth.

There are some parameters of methane oxidation that can be affected by environmental factors such as temperature, water content, nutrients, soil type and oxygen concentration (18, 27 and 39).

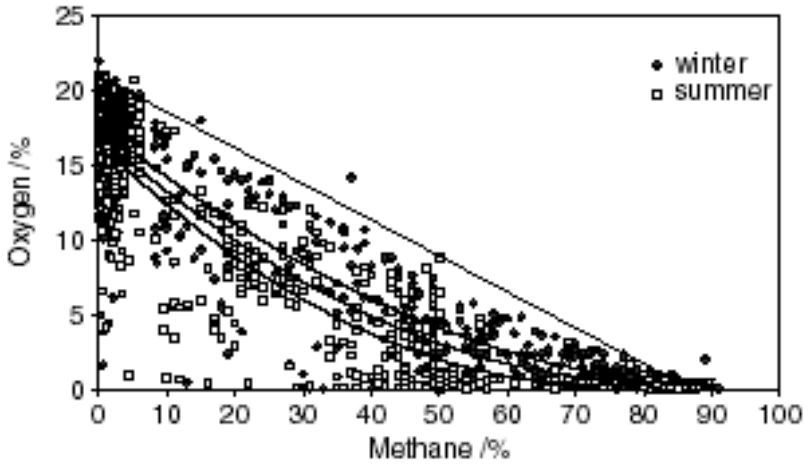

Figure 2. Soil oxygen expressed as functions of soil methane. The straight line represents the relationship expected with simple displacement. The fitted curves represent averages (from bottom to top, respectively) of summer and winter (27)

From the Figure 2 above, the straight line the oxygen intercept was 21 %, this correspond to the atmospheric value. Methane will displace in soil air by natural gas is 80-95%. From the curve, the intercept is estimated as 87.5%

The Figure 2 shows that there is a strong indication of methane oxidation done by methanotrophic bacteria. Oxygen depletion measured as the difference between the measured oxygen and the value after displacement, is about one-third greater in summer than in winter.

It was discovered that the lack of oxygen in the soil was partly caused by the oxygen being replaced by the gas, but was largely the result of the gas in the soil being consumed by bacteria that oxidized the gas and thereby removed oxygen from the soil (1).

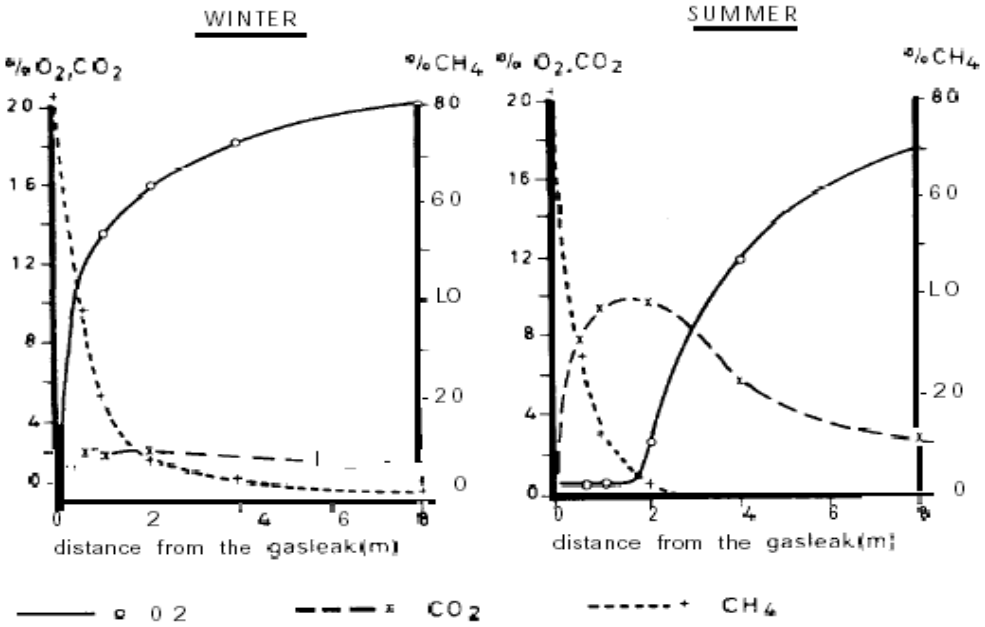

Figure 3. The influence of the season on the composition of soil air around an artifial gas leak of 25 l/h (19)

It may be concluded from this study that methanotrophic bacteria act as consumer of methane in aerobic soils with a significant oxidation of methane concentration during gas leaks in the soils. In the surroundings of natural gas leaks, methane, ethane and possibly some other components of the natural gas are oxidized by microbial activities as long as oxygen is available. This would be demonstrated by an increased oxygen consumption and carbon dioxide production.

METODE

This study was done to evaluate the modification of composition induced by bacterial activity in order to compare with bibliography and field condition in Research Department Gaz de France by flowing a continuous injection of gas in the top soil where the micro organisms live. Thus we suppose that gas will spread out in to the soil and the methane coming from natural gas will be degraded in the soil. To describe the location of sampling area. It shows that at the point 1 is the source of the injection. So, this point does not need to be measured.

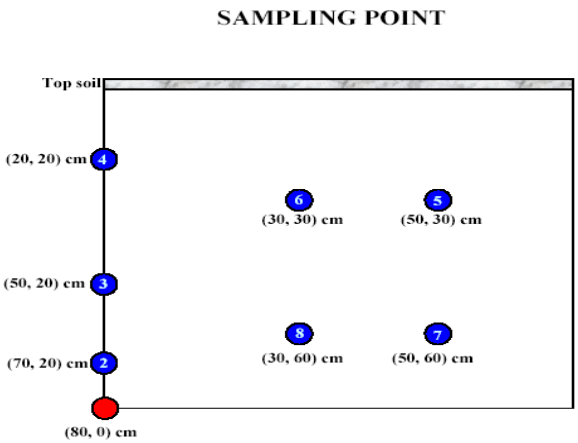

Figure 4. Stimilation of gas leak in the top soil

The installation described in Figure 4 is a stimulation of a real leak underground. Gas is injected continuously through the injection tube and diffuses underground as with a real leak. The inserted tubes 1 to 8 are used for sampling the atmosphere at the location where the tube extremely is located. A sample is taken at location 0, far from the leak in order to have the atmosphere composition such as N_2, O_2 and CO_2 without the influence of natural gas. The composition of each sample (2 to 8) is analysed such as N_2, O_2, CO_2, CH_4 and other hydrocarbons. The gas composition and particularly its methane content are known a gas chromatograph is installed in a laboratory not far from the location where this experiment is taking place for general purpose.

The sample 0 is considered as reference and compared with the sample 2 to 8. For each sample the concentration of O_2, CH_4 and CO_2 are evaluated considering that the sample composition is the result of the mixing of gas and air with the same composition as that of sample 0. Then these theoretical compositions are compared to the composition coming from each sample analysis. The deficit of O_2 and CH_4 with the enrichment of CO_2 is considered as an indicator of methanotrophic bacteria activity.

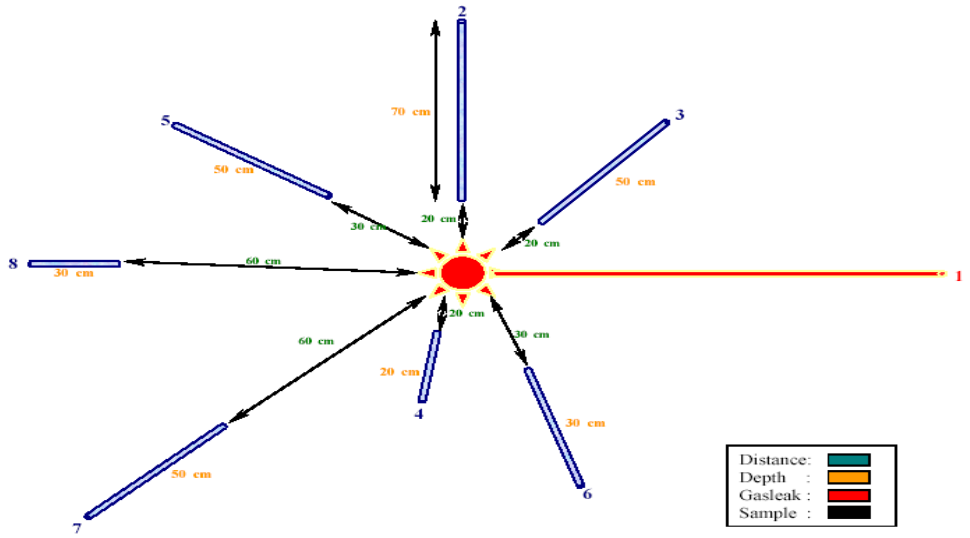

Figure 5. Sampling Area

All top of those tubes (except injection tube) should be closed in order to avoid gas spreading out from the ground through then. One tube is linked to the gas supply into a building close.

To be able to control the debit of the gas leaks injected, it should be connected by a debit meter that can control the debit during the study.

In this study, we use 10 l/h for the debit control, and this instrument should be checked everyday in order to see the stability of the gas leaks itself.

2.1 Gas Sampling

It was used a gas detector CATEX+. The CATEX + is an explosimeter-catharometer with three measurement scales: ppm, LEL and volume gas. It is used to pinpoint the exact location of a leak. It can be operated in both indoor and outdoor hazardous atmospheres.

The small pipes injected in the soil was connected to this apparatus by holding an ampoule (gas sampling bottle) to detect the concentration of methane spread out from source leak. These samples then were analyzed in laboratory used gas chromatography to separate an organic compound.

2.2 Gas Chromatography (GC)

Gas chromatography is an instrument that can be used for separating an organic compound that is volatile. It consists of a flowing mobile phase, an injection port, a separation column containing the stationary

phase, a detector and a data recording system (software system). The way to separate an organic compound is by partitioning between the mobile gas phase and the stationary phase in the column.

Micro GC Agilent 3000 allows measurement of most compounds from methane to hexane, of natural gas and permanent gases (H_2, O_2, N_2, CO, CO_2, He, Ar). SOPRANE is software making it possible to ensure the complete management of an analyzer and its peripherals.

2.3 Comparison bacterial activity in the soil theoretically and practically

To determine the methane consumed by methanotrophic bacteria in the soil, we need to compare the concentration of methane, oxygen and carbon dioxide that we have in each sample 2 to 8 with those that would be give the result from natural gas mixed with soil air.

To determine the theoretical value, we use the data from point 0 which is full of soil air. We use the following notation for the equation. We assumed that the total amount of air in the soil is equal to 1 (one). We use the subscript 0 (zero) for those components in the soil air where there is no gas leak injected in the soil as a reference sample.

So,

$[O_2]_0 + [CO_2]_0 + [N_2]_0 = 1$

We assume that the other components in the soil air such as; argon, helium, etc are the very small components, and we neglect them.

So, for the points where there is natural gas injected in the soil, the component of gasses in the soil become:

$[CH_4 + C_2H_6 + C_3H_8 + C_4H_{10} + C_5H_{12}]_d + [O_2]_d + [CO_2]_d + [N_2]_d = 1$

The subscript 'd' mean the location where there the samples are taken.

Although the methanotrophic bacteria prefer to consume CH_4 than other alkenes components, we need to use the value of other alkenes that we found during measurement by using gas chromatography. Because the chemical compositions of natural gas do not only consist of methane, there are still other alkenes compositions in it. The next primary chemical reaction shows the general formula for oxidation (19):

$CH_4 + 2\,O_2 \rightarrow CO_2 + H_2O$

Naturally, if there is only natural gas displacing the soil air, in this case, the proportion of gasses in it should be constant. From each samples, we assume that the concentration of gasses is the result of mixing of a fraction "x" of soil air and a fraction "y" of gas. If the gas leak spread out in the soil, it means that, the composition of gas will mix together with soil air. So, we can say x+ y=1.

To calculate the value of x and y at a given location, we assume that nitrogen is only displaced by natural gas.

Thus at any location $x = \frac{[N_2]_m^d}{[N_2]_m^0}$

Where "d" indicates a concentration measured at any location (points "2 to 8") and "0" indicates the value measured on the point 0 as a reference.

Thus, the theoretical oxygen concentration is related to:

$$\frac{[O_2]_{th}^d}{[O_2]_m^0} = \frac{[N_2]_m^d}{[N_2]_m^0}$$

The gas concentration should be

$y = 1 - x$

Thus the methane concentration in "d" should be

$$[CH_4]_{th}^d = (1-x)[CH_4]_{inj}. \quad (1)$$

$[CH_4]_{inj.}$ Is the methane concentration of the gas injected in the ground, this is taken as the average methane concentration of the gas injected.

Furthermore, if we assume that higher alkanes are not consumed by bacteria, then the amount of $[C_2^+]$ (total alkanes except methane) at a given location represent the molar fraction "y" of gas injected through the relationship of:

$$y = \frac{[C_2^+]_m^d}{[C_2^+]_{inj}} = (1-x) \quad (2)$$

From the equation 8, we can check the consistency of the measurements by comparing the value of $[C_2^+]$ measured to that deriving from the nitrogen concentration.

So from that formula, says that :

$$[C_2^+]_m^d = [C_2^+]_{th}^d = (1-x)[C_2^+]_{inj}$$

Theoretically, the amount of CH_4 consumed ($[CH_4]_c$) by methanotrophic bacteria is:

$[CH_4]_c = [CH_4]_{th} - [CH_4]_m$ ..... (3)

Where :

$[CH_4]_{th}$: CH_4 component theory

$[CH_4]_m$: CH_4 component measured

If we look at the chemical reaction between methane and oxygen, we know that 1 mol of methane needs 2 mol of oxygen to have an oxidation reaction.

According to that reaction, 2 mol of $[CH_4]_c$ would be oxidized 1 mol of $[O_2]_0$.

So, we could conclude that;

$$[O_2]_m = [O_2]_{th} - 2[CH_4]_c \quad (10)$$

Where :

$[O_2]_m$: O_2 component measured

$[O_2]_{th}$: O_2 component theory

$[CH_4]_c$: CH_4 component consumed

For carbon dioxide that will produce by oxidizing reaction:

$$[CO_2]_p = [CO_2]_{th} + [CH_4]_m \quad (11)$$

Where :

$[CO_2]_c$: CO_2 component produced

$[CO_2]_{th}$: CO_2 component theory

$[CH_4]_m$: CH_4 component measured

For each sampling point the difference of oxygen, methane and CO_2 concentration are calculated by comparing the theoretical concentration calculated as above and the real values as measured in each sample by using gas chromatography. These differences are expressed by

$$\Delta_{CH_4} = [CH_4]_m - [CH_4]_{th} \quad (12)$$

$$\Delta_{C_2^+} = [C_2^+]_m - [C_2^+]_{th} \quad (13)$$

$$\Delta_{O_2} = [O_2]_m - [O_2]_{th} \quad (14)$$

$$\Delta_{CO_2} = [CO_2]_m - [CO_2]_{th} \quad (15)$$

Where:

Δ_{CH4} : difference value CH_4 between measurement data and theory

Δ_{CH4} : difference value CH_4 between measurement data and theory

Δ_{O2} : difference value O_2 between measurement data and theory

Δ_{CO2} : difference value CO_2 between measurement data and theory

$[CH_4]_m$: CH_4 concentration measured

$[CH_4]_{th}$: CH_4 concentration theory

$[C_2^+]_m$: total alkenes except CH_4 concentration measured

$[C_2^+]_{th}$: total alkenes except CH_4 concentration theory

$[O_2]_m$: oxygen concentration measured

$[O_2]_{th}$: oxygen concentration theory

$[CO_2]_m$: carbon dioxide concentration measured

$[CO_2]_{th}$: carbon dioxide concentration theory

RESULT AND DISCUSSION

The composition of the $[C_2^+]$ gives an indicator of the consistency of the measurement. The following Figure 6 to Figure 12 show the difference between O_2, CO_2, CH_4 and C_2+ .

Thus positive values mean that there is an excess of gas in the sample and a negative value is a lack of gas in the sample if we compare with the values coming from mere displacement.

From those figures, we found there is lack of O_2 that means the measured concentration of oxygen is lower than the value expected from displacement. On the other hand, we found there is excess in carbon dioxide and methane that are greater than the value expected from displacement.

All the following figures clearly show the theoretical concentration of oxygen is lower than measured data, and on the contrary, the concentration of carbon dioxide is higher than measured data. Then, the concentration of alkanes except methane is stable and consistent. It means that, there is no oxidation reaction of methane to produce CO_2.

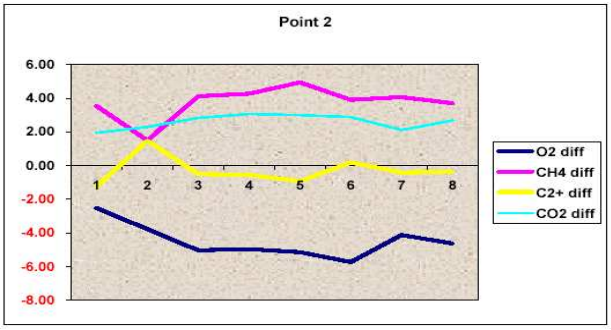

Figure 6. The average difference value between O2, CO2, CH4 and other alkanes at point 2

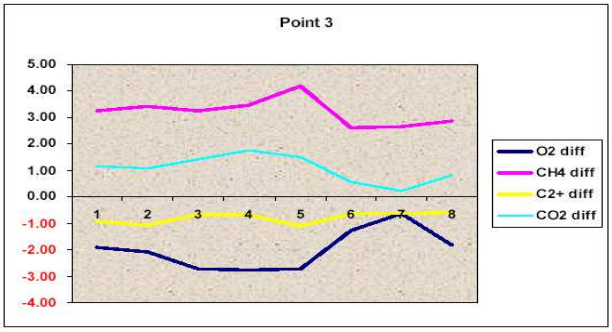

Figure 7. The average difference value between O2, CO2, CH4 and other alkanes at point 3

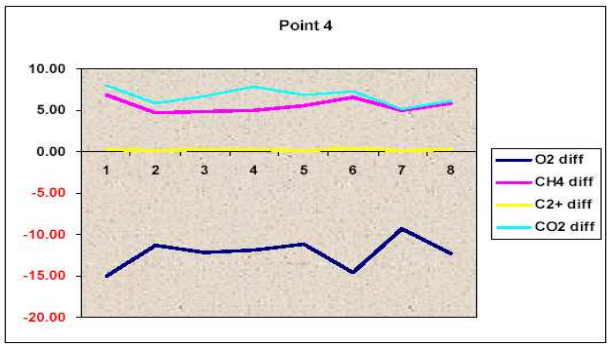

Figure 8. The average difference value between O2, CO2, CH4 and other alkanes at point 4

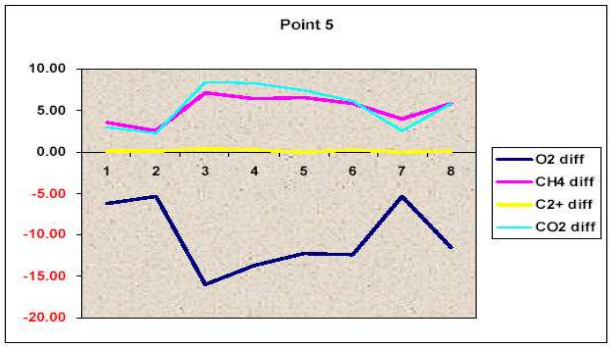

Figure 9. The average difference value between O2, CO2, CH4 and other alkanes at point 5

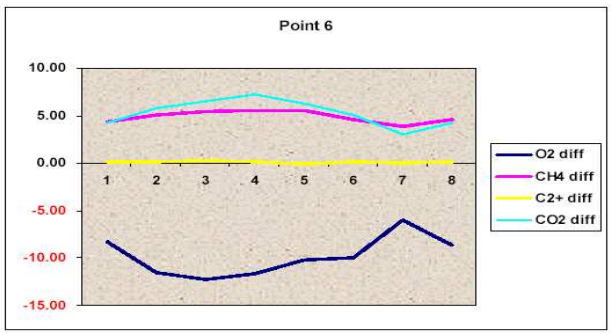

Figure 10. The average difference value between O2, CO2, CH4 and other alkanes at point 6

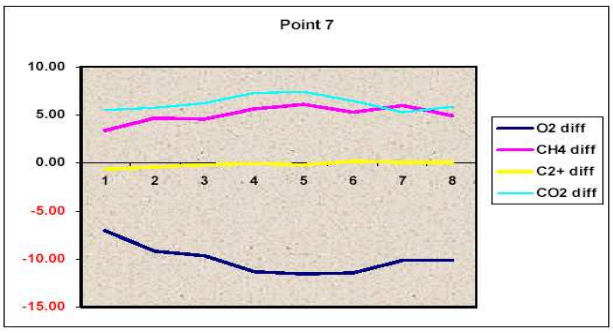

Figure 11. The average difference value between O2, CO2, CH4 and other alkanes at point 7

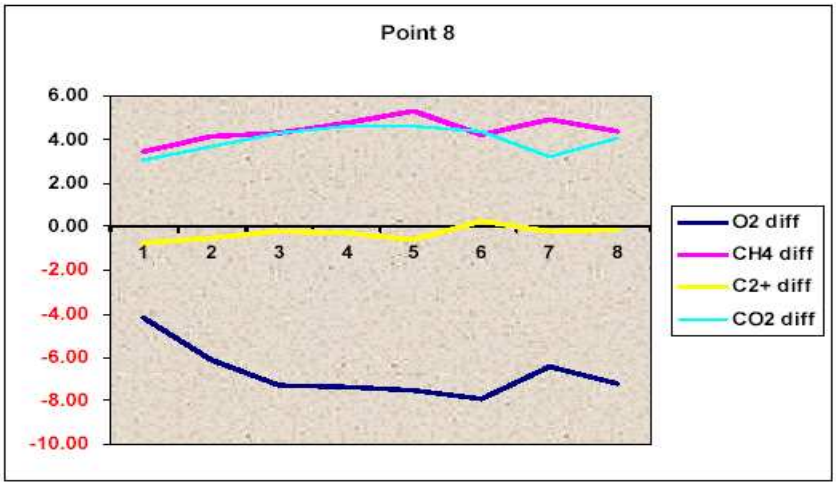

Figure 12. The average difference value between O_2, CO_2, CH_4 and other alkanes at point 8

Figure 13 showed the reduction of methane in sampling points compared to the leak source. Closer to the leak source higher the methane losses.

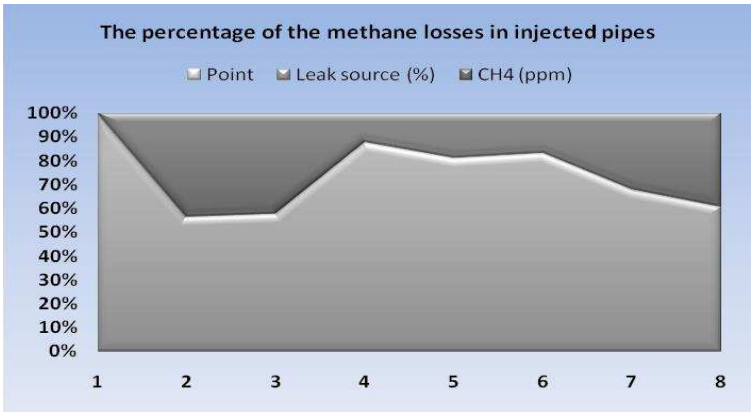

Figure 13. The percentage of methane losses in all sampling points

CONCLUSION

1. The measurement shows that there was a deficit of oxygen that can reach into 10% (v/v) and an excess of methane and CO_2 of about 5%.
2. The micro gas chromatography data was found that the pipes injected from 20 cm leak are 77.16% CH_4 loss in 70cm depth, 73.15%

in 50cm depth and 14.08% in 20cm depth. And the pipes injected from 30 cm leak are 20.27% in 30 cm depth and 65.13% in 60 cm depth. Then, the pipes injected from 50 cm leak are 23.40% in 30 cm depth and 47.40% in 60 cm depth. The leak source is in 80 cm depth.

3. Significant losses of methane were recorded in injected pipes closed to the leak source (such as the pipe injected on 50 cm, 70 cm in 20 cm distances and 60 cm depth in 30 cm distance).

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