Literature review: Effect of gas leakage on vegetation

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date: 8.5.2025

1. Methane effect on plants

Methane is not a toxic gas; its adverse effects are indirect. As a result of a natural gas leak, methane concentration in the soil increases, which leads to a decrease in oxygen content through two mechanisms. 1. Methane directly displaces vital oxygen (an inverse relationship between methane and soil oxygen). 2. Oxygen is consumed by methanotrophs (methane-oxidizing bacteria) to break down methane [1]. For example, it has been found that up to 90% of methane is oxidized at rates as high as 230 g/m^2 of soils per day in summer. *Methylocystis* and *Methylocella* are thought to be key bacteria in this process [2]. Methane oxidation was found to occur much faster in mineral-rich soil [3]. Oxygen depletion causes anaerobic soil conditions that can significantly limit respiration and growth of plant roots, especially in the early stages of development. In turn, anaerobic stress can also reduce plant root resilience to other factors such as pests and diseases, which contribute to overall vegetation decline and death. As a result of natural gas leaks, vegetation death is caused by a shortage of oxygen (hypoxia) or a total absence of oxygen (anoxia), which rather quickly leads to death by suffocation. The extremely low oxygen concentration is regarded as the most important cause of death of plants. Plants are obligate aerobic organisms and cannot survive for long periods in low-oxygen environments.

2. Hydrogen effect on plants

The mechanisms of the harmful effects of methane and hydrogen on plants may be similar.

Like methane, hydrogen is non-toxic and, therefore, it does not directly harm plants. But H_2 is also consumed by soil microbiota — hydrogen-oxidizing bacteria (aerobic and anaerobic HOB). Aerobic HOB use hydrogen as an electron donor and oxygen as an electron acceptor. This process can also cause oxygen depletion and anaerobic soil conditions, especially in poorly-aerated clay-rich soils, where H_2 diffusion is slow and H_2 transport is impeded.

In well-aerated soils, the influence of small gas leaks on plants can be not significant. For example, in aerated meadow soils along Italian Alps valleys, the concentrations of H_2, CH_4 and CO_2 reach 1, 51 and 27 vol%, respectively. Despite the extraordinarily elevated concentrations of these gases, vegetation inhibition is not observed [4]. Moreover, 1 vol% of H_2 can positively affect pants. H_2 is known to be an obligate byproduct of nitrogen fixation in legume rhizobia. For 200 kg of fixed nitrogen per hectare of legumes there are 240,000 liters of hydrogen [5].

H_2 is a gaseous signaling molecule, powerful antioxidant, anti-inflammatory, and anti-apoptotic agent. The concept of “H_2 fertilization” has become a milestone in the advancement of hydrogen agronomy. Hydrogen can significantly enhance plant resistance to different types of environmental stresses. Hydrogen effectively mitigates temperature (cold and heat), light (UV-A, UV-B), heavy metal (Hg, Cd, Al), pesticide (PQ), salt, drought and osmotic stresses. H_2 improves the antioxidant and photosynthetic capacity of plants, enhances enzyme activity, regulates the expression of antioxidant and ion transporter genes, reduces reactive oxygen species (ROS) accumulation and lipid peroxidation (LPO), inhibits effects of metal pollution, reduces heavy metal accumulation, maintains ion homeostasis, regulates sugar metabolism, increases plant resistance against disease (tolerance towards biotic stresses), enhances lateral root and adventitious root formation, promotes the growth of beneficial microorganisms and soil fertility, improves the quality of vegetables and fruits. In plants, H_2 acts as an important bio-regulator that modulates various physiological processes from seed germination to post-harvesting [5, 6].

In the forms of hydrogen-rich water (HRW) and H_2 gas, H_2 is the protagonist of hydrogen agronomy. Generally, the concentration of dissolved H_2 in HRW utilized in agriculture is about 78 µmol/L (10% HRW) [7] and the H_2 content during fumigation tests is greater than 0.2 µmol/L [8]. The application of HRW is believed to be an effective and safe method to exert the biological effects of H_2 [5]. The maximum molar solubility of H_2 in water under atmospheric pressure and at room temperature is 0.78 mM (1.56 mg/L) [9]. Regular 100% HRW contains 0.78 mmol of H_2 per 1 L. The upper limit of the beneficial antioxidant and bio-stimulating effect of hydrogen on plants must be determined experimentally.

Despite the important biological role of hydrogen, its high subsurface/surface concentrations and long-term exposure definitely lead to vegetation suppression and disappearance, as evidenced by the sites of geological seeps hydrogen seeps (approximately 10 – 60 vol% H_2, so-called golden hydrogen). Natural emissions of hydrogen create topographical circular or elliptical depressions and gaps in vegetation. These barren patches are often referred to as "fairy circles" (although some of them are not related to H_2 escape). Importantly, concentrations of H_2 within soils in these superficial anomalies typically range between 10 and 1000 ppmv (0.001 – 0.1 vol%), the highest concentration being 8000 ppmv (0.8 vol%), which highly suggests a biological origin for the hydrogen hotspots, e.g., fermentation in wet soils, nitrogen fixation, and cellulose decomposition by termites etc. In case of massive geological seeps from sedimentary basins, H_2 concentrations should be much higher and H_2 flux should be driven by advection (pressure gradients, Darcy's law) [10].

The key point, regardless of the origin of the H_2 sources (advective geological seeps with high concentrations, modern microbial production with low concentrations or possible artificial – pipeline leakages), is that strong and prolonged hydrogen exposure results in gaps in vegetation that are visible to the naked eye. Although gaps in vegetation may not be caused by direct hydrogen exposure, but by a combination of factors or accumulation of water in depressions (waterlogging, shallow aquifer).

In a long-term project to detect hydrogen leaks (GetH2), vegetation has been cited as a hindrance to their detection since plants are capable of reflecting fluorescence signals and distorting measurement results obtained using a laser and the effect of inelastic light scattering (the Raman effect) [11].

Hydrogen vs Methane (physical and chemical properties)

Cooling effect caused by decompression

During throttling or decompression, almost all gases experience significant cooling, known as the Joule-Thomson effect. This is a temperature change phenomenon that occurs when a gas is allowed to expand adiabatically (without heat exchange with the surrounding space) from a region of high pressure to a region of extremely low pressure. The inversion temperature of most gases is above ambient temperature (methane – 680°C or 953K), but for hydrogen the inversion temperature is about -73°C or 200K. Below the inversion temperature, a non-ideal gas (any gas) expanding at constant enthalpy will experience a decrease in temperature, and above which, an increase in temperature.

During decompression, hydrogen will experience heating.

Highly compressed hydrogen can ignite when throttled, this must be taken into account, since hydrogen seeps very well through the smallest pores and even through some materials.

A numerical study showed that the Joule-Thomson coefficient decreases by about 30% and 50% at hydrogen mole fractions of 15% and 30% in hydrogen-blended natural gas, respectively [12].

Under standard conditions the density of hydrogen (*p* = 0.0838 g/L) is 8 times less than that of methane (*p* = 0.668 g/L), which ensures greater buoyancy of hydrogen. In other words, due to the much lower density, hydrogen will migrate vertically faster, rising to the surface.

Moreover, compared to natural gas, hydrogen has a smaller viscosity (approximately by 15%), which results in a stronger convective effect. According to various literary sources, the diffusion coefficient of hydrogen is 2.8 to 6 times higher than that of methane [13-15]. Compared to natural gas, hydrogen exhibits faster pressure and velocity decay. The pressure of natural gas and hydrogen leaking at the ground surface is less than 1 Pa and 0.1 Pa, respectively. At any given moment, the diffusion range of hydrogen is significantly larger than that of natural gas [14].

Hydrogen forms an "n" shaped distribution while methane forms an "M" shaped distribution, with higher hydrogen concentrations observed further from leakage holes [13].

Similar to natural gas, hydrogen distribution in soil can be categorized into three stages [14, 16]:

1) rapid growth (or the initial leakage stage controlled by advection caused the pressure gradient),
2) slow growth (or the diffusion stage caused by the molecular concentration gradient),
3) stabilization.

Compared to natural gas leakage, the growth stage of hydrogen lasts longer, with natural gas diffusion reaching stability around t = 6 min, while hydrogen reaches stability at approximately t = 9 min. Hydrogen diffuses rapidly in soil, with the time for hydrogen reaching the lower explosive limit (LEL) at the ground surface approximately eight-times faster than natural gas. Numerical modeling shows that under the same conditions, it takes natural gas 43.18 min to reach the lower explosive limit at the earth's surface, whereas hydrogen has already reached its LEL at a depth of 45 cm underground in about 5.47 min [14]. The explosive limits of hydrogen and methane are 4%-75% and 5%-15%, respectively [17].

Undoubtedly, the diffusion of hydrogen in the soil occurs much faster compared to natural gas diffusion, which makes hydrogen leakage are much more dangerous. In addition, hydrogen is much more aggressive towards metals. The small size of hydrogen molecules and high diffusivity allow it to leak into materials, a process known as **hydrogen embrittlement**. Hydrogen can easily attack metals (various types of steel). It gradually penetrates the metal structure reducing its strength and making it more susceptible to cracks and leaks [11, 18].

Effect of hydrogen-blended ratio (HBR)

Given the extensive natural gas pipeline infrastructure, blending hydrogen with natural gas and transporting it through existing natural gas pipelines could be the most viable approach to achieving large-scale hydrogen transportation [14].

The method of gradually adding hydrogen to natural gas is used in many developed countries (France, Germany, Italy, Norway, Netherlands and Britain) with the current safety limit not exceeding 50% [19].

Given the aggressiveness and corrosiveness of hydrogen, the gradual addition of hydrogen to natural gas is technically justified. In many numerical simulations, the influence of hydrogen ratio on the properties of hydrogen-blended natural gas (HBNG) rather than pure hydrogen is very often considered.

The first dangerous time* (FDT) for H_2 ratio of 0%, 5%, 10%, 15%, and 20% were 43.18 min, 37.95 min, 36.4 min, 35.03 min, and 33.47 min, respectively (Table 1). Moreover, adding H_2 decreases HBNG density and viscosity, enhancing the gas diffusion and reduces the LEL of HBNG (Table 1) [14].

It is noteworthy that the greatest reduction (gap) in the FDT was between 0% and 5% HBR, the FDT was shortened by 5.23 min. The subsequent increase in hydrogen concentration from 5% to 10%, 15%, and 20%. led to a gradual reduction in the FDT by 1.55 min, 1.37 min, and 1.56 min, respectively [14].

In general, the FDT for HBNG leakage is substantially shorter than for natural gas, emphasizing the need for timely leak detection and response. Increasing the HBR accelerates the diffusion process and decreases the FDT, posing greater risks for pipeline safety [14].

Different hydrogen ratios (HBR) have a certain effect on the diffusion range in the vertical direction and have less effect in the horizontal region [19].

Table 1. Effect of hydrogen concentration on the lower explosion limit (LEL) and the first dangerous time (FDT) for hydrogen-blended natural gas (HBNG)

Hydrogen Blending Ratio (HBR)	LEL	FDT
0	5%	43.18 min
5%	4.9383%	37.95 min
10%	4.878%	36.4 min
15%	4.8193%	35.03 min
20%	4.7619%	33.47 min

The LEL of pure H_2 is 4%.

**The first dangerous time is the time required for the gas concentration at the surface projection point of the leakage hole to reach the lower explosive limit (LEL) [14].*

Laser detection of gas leakages

Using a laser measurement system, it is much easier to detect leaks of methane than those of hydrogen, since CH_4 has characteristic absorption lines in infrared (IR) light (3.5 and 8 microns). When IR laser light hits a methane cloud on the ground, some of it is absorbed by the molecules and significantly weakens the light of that wavelength. This method is not applicable to hydrogen, because it has extremely low absorption. Hydrogen is very difficult to measure. The Raman effect (inelastic light scattering) can be used to detect hydrogen leaks. Hydrogen scatters light of a certain wavelength (e.g., 532 nm) back with slightly lower energy. This is a rather expensive, complex and cumbersome method in practice (a measuring system that fits into a helicopter must weigh no more than 100 kilograms) [11].

Hyperspectral remote detection of vegetation affected by natural gas micro-leaks

Most studies using multispectral or hyperspectral imaging are based on the fact that plants absorb and reflect certain ranges of the electromagnetic spectrum. Healthy plants are known to actively absorb red light (650–680 nm) and reflect near-infrared (NIR, 785–900 nm) light. Plants with a higher chlorophyll content reflect more near-infrared energy (50%) compared to unhealthy or dead plants (40%). The normalized difference vegetation index (NDVI) is based on this phenomenon. NDVI is calculated as (NIR - R) / (NIR + R) and ranges between -1 and 1, where values of -1 – 0 correspond to dead plants or objects and 0.66 – 1 indicate very healthy dense vegetation. All remote methods of monitoring gas leaks using plants are somehow related to the above-mentioned phenomenon and index (its modifications and improvements).

This technique can be applied to plants exposed to both beneficial and harmful effects of hydrogen leaks.

Beneficial effect: under the influence of hydrogen, photosynthesis is enhanced. An increase in the number chloroplasts under soil degassing conditions leads to changes in the optical characteristics of plants. (Based on the many biological effects of hydrogen and the novel concepts of “hydrogen fertilizer”, “hydrogen agronomy and agriculture”, it is possible to conclude that H_2 micro-leakages may have a positive effect on the vegetation in well-aerated sandy soils.)

Harmful effect: under the influence of hydrogen, photosynthesis is slowed down. A decrease in the number chloroplasts under soil degassing conditions leads to changes in the optical characteristics of plants.

Various indices have been developed for the timely detection of methane leaks using spectral analysis of plants: modified chlorophyll absorption reflectance index (MCARI) with commendable sensitivity but limited separability in recognizing stressed grasses; optimized soil-adjusted vegetation index (OSAVI) that with higher sensitivity and separability in early detection of methane leaks. OSAVI was able to discern vegetation stress 21 days after the onset of methane exposure. Deep Neural Networks detected methane leaks after three weeks of methane exposure with 98.2% accuracy. DNN results indicated an increase in visible (VIS) and a decrease in near-infrared (NIR) spectra due to methane exposure [20].

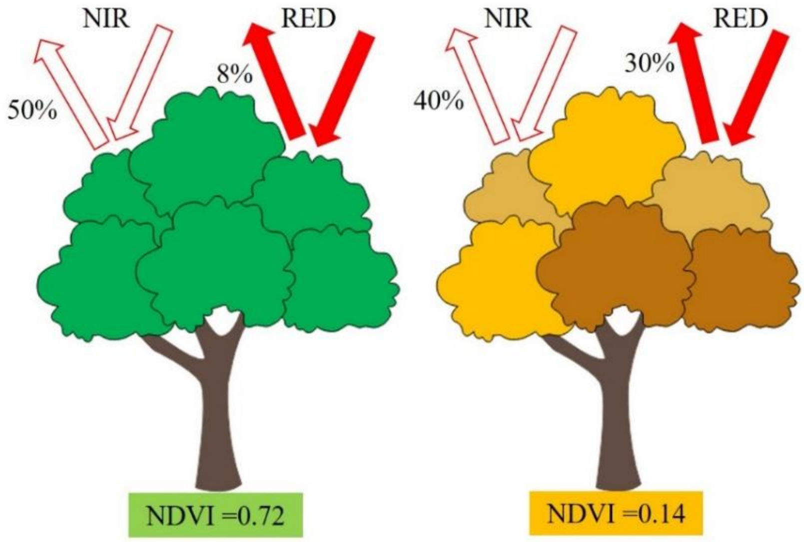

Fig. 1. Spectral difference between a healthy and an unhealthy plant and NDVI values
The upward arrows indicate spectral reflectance of vegetation, downward – light absorption

In a study of natural gas pipeline micro-leaks using optical plant characteristics, the 616 nm (Orange) and 829 nm (NIR) bands were found to be most sensitive to natural gas stress for the three plant species. The variational mode decomposition index (VMDI) could identify stressed wheat and grass one week earlier than other indices and could better detected stressed vegetation throughout the phenological cycle [21].

Spectral remote detection of vegetation affected by hydrogen leaks is possible with certain adjustments to indices and spectra.

Effect of soil properties on leaks of natural gas (predominantly CH_4)

The data presented are based on a series of controlled field-scale experiments [22] and actual natural gas leaks [23].

Despite the extreme importance of soil factors (texture, permeability, moisture, saturation), their influence on the behavior, distribution and accumulation of methane must be considered in conjunction with the relative contributions of advection and diffusion, which vary with the distance from the leak source (Fig. 2). Soil properties will influence both advection and diffusion by increasing or decreasing them. Differences in soil texture and moisture definitely complicate the behavior and migration of gases and, therefore, the assessment of leakage rate and grade (classification). Moreover, the influence of soil properties will not always be unambiguous over time and with changing weather conditions.

Relative contributions of advection and diffusion

Advection and diffusion are the main mechanisms controlling methane transport in the soil in addition to biological activity (e.g., microbial oxidation). Horizontal and vertical migration of CH_4 occurs because advection, induced gas release (pipeline pressure), plays a primary role close to the leak point. In the area close to the leak point, the advective flux is much larger and stronger than the diffusive flux, especially in the vertical direction (in the vertical direction, the concentration contours are convex upward, meaning that methane is predominantly moving upward ↑). As the distance from the leak point increases (e.g., 1.5 m away for the field test bed and 5 m away for the actual gas leakages), the advection effect caused by the pipeline pressure (Darcy's law) gradually decreases to a minimum value and its dominance disappears. The diffusion effect caused by the molecular concentration gradient (Fick's law) is weaker than the advection effect controlled by the pressure gradient. The relative advection and diffusion contributions to methane behavior and migration depend on a distance from the leak point [22].

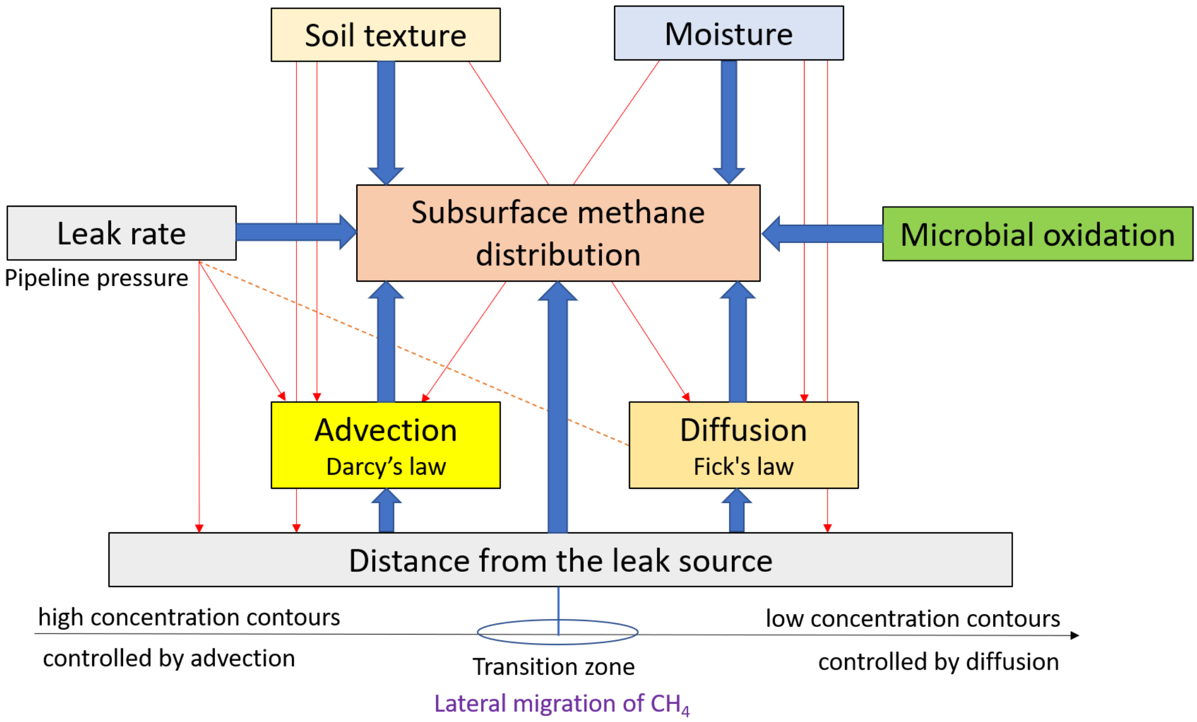

Fig. 2. Combination influence of soil properties, leak rate and distance, advection and diffusion (The dashed line shows that leak rate has little effect on diffusion.)

CH_4 concentrations typically decreased exponentially with increasing distance from the leak site. A larger concentration gradient is observed in the area farther from the leak point, represented by dense contours. Approaching the leak point, the high concentration contours are more sparsely distributed. Advection dominates natural gas transport up to a certain distance and drives the distribution of higher concentration contours. Beyond this distance, diffusion dominates the distribution of lower concentration contours to the far-field. Although large leak rates (0.5 kg/h and 0.85 kg/h) initially lead to faster and further gas migration, the leak rate has little influence on the diffusion dominated migration farther from the leak source [22].

Soil texture

Soil texture (related to many other soil properties: water infiltration, water-holding capacity, aeration etc.) plays a very important role in the behavior and migration of methane.

Gas transport, initially controlled by advection and then by diffusion, is significantly complicated by differences in soil permeability (texture) as well as competition between vertical and horizontal flows in the vicinity of the leak point.

Advection. The advective effect and transport will be stronger in highly permeable sandy soils, resulting in faster gas flow at higher concentrations through the high permeability pathways near the leak point, where methane predominately migrates upward and escapes to the atmosphere. Thus, the volumes of gas further moved and controlled by diffusion will decrease. Accordingly, the overall methane spreading in sandy soils is often less than in clay soils. Conversely, in low permeability clay soils the advective effect and transport will be less, resulting in slow but length scale migration due to diffusion.

Diffusion. Diffusion rate directly depends on soil permeability: it is highest in sand and lowest in clay. Due to the slow diffusion rate, the overall gas distribution in low permeability clays is often greater than in highly permeable sands. The more predominant impact of the gas diffusion in low permeable soils affects the gradual spreading width of the CH_4 plume, resulting in more outstretched contours. Diffusive behavior is also observed in highly permeable soils but to a lesser extent due to the competition with vertical gas transport to the soil surface and gas escape to the atmosphere [22, 23].

Soil homogeneity and heterogeneity

Since the permeability of clay (3.82E-14 m^2) is over three orders of magnitude smaller than that of sand (2.90E-11 m^2), in inhomogeneous soils gas preferentially migrates around the clay layers. The presence of the high-saturation clay layers within the soil leads to different gas migration behavior compared to the homogenous soil conditions. Locally higher methane concentrations (e.g., hot spots) may be detected adjacent to a low permeable clay layer. Hot spots may continue even after a leak is repaired due to the accumulated gas underneath the low permeable soil layer. This indicates the importance of understanding the soil layering conditions during leak survey and how that layering can lead to subsurface gas distributions and transport [22].

Soil moisture content

Gas spreading, initially controlled by advection and then by diffusion, is also complicated by increased soil water saturation and impeded gas transport in the areas of high-moisture content.

The water-holding capacity of soil depends on its texture.

In general, the effect of soil moisture on diffusion is inversely proportional: the higher the soil moisture (and also tortuosity induced by water), the weaker the diffusion. However, the influence of soil moisture on advection is much more complex. On the one side, the effect of soil moisture on advection should be also inversely proportional because of reducing gas permeability: the higher the soil moisture, the less the gas permeability and the weaker advection. But, on the other hand impeded gas transport in the high-moisture content areas may cause local pressurized CH_4 accumulation and enhance the advection effect near the leak source. That is, the influence of soil moisture on advection can be described as directly proportional, but up to a certain distance (advection attenuation and diffusion domination).

Preliminary numerical [22] and then actual field studies [23] confirmed that water saturation has an inverse effect on the contributions of advection and diffusion to CH_4 distribution. Higher soil water saturation leads to a stronger advective flux but a weaker diffusive flux. The higher soil water saturation increases the tortuosity and pressure for gas flow, thus increasing the advection while at the same time lowering the lateral diffusion.

The study, based on 199 soil samples collected from June 2019 to March 20207 at 77 actual pipeline leakages of natural gas, found that given the same soil texture (due to standardized pipeline backfill protocols), soil moisture has a dominant effect on methane behavior, concentration and migration range. High soil moisture content resulted in reduced lateral diffusion due to water-induced tortuosity and, therefore elevated concentrations near the leak point. Lateral CH_4 migration was assumed to be dominated by diffusion, starting at 5 m from the leak sites, whereas transport within the immediate vicinity of the leak was controlled by advection [23].

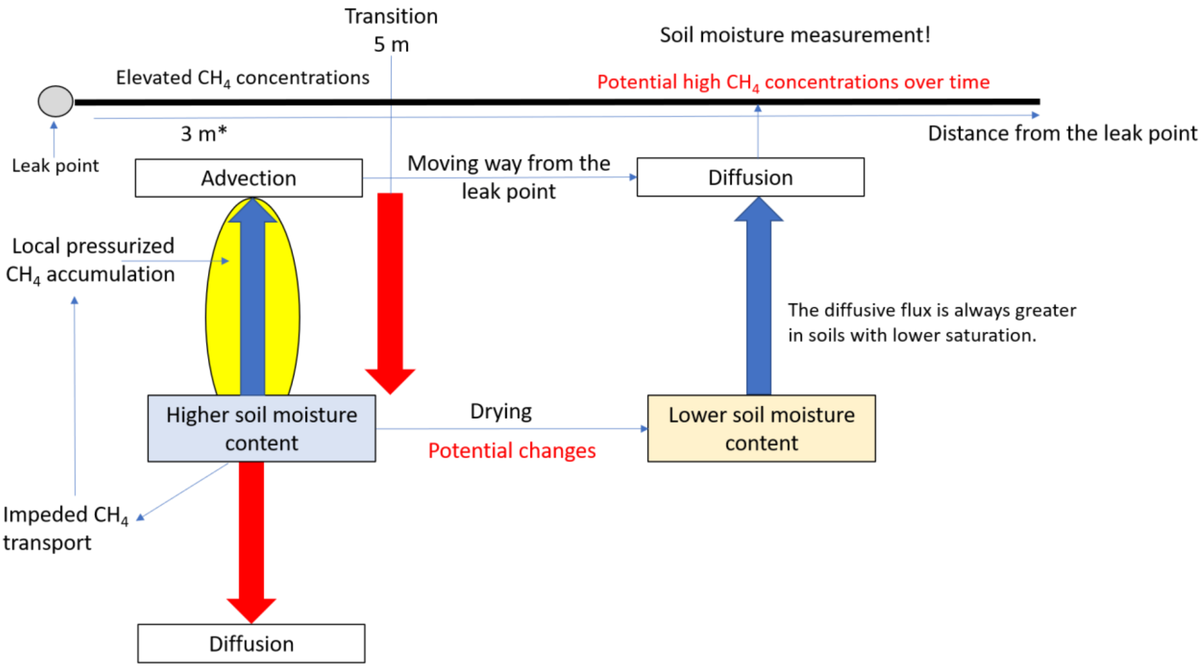

Fig. 2. Soil water saturation effect on the contributions of advection and diffusion to CH_4 distribution depending on distance from the leak point (blue arrows indicate increasing advection and diffusion, red – decreasing)
* – 82% of the subsurface gas concentrations exceeding the LEL (5 vol% or 50,000 ppmv) were detected within 3 m of the actual leak location [23]

The importance of measuring soil moisture during gas leaks

As the soil dries, methane diffusion will increase, and therefore, the distance of elevated gas concentrations may increase. Therefore, to avoid misclassification of leaks, the inspector (surveyor) should record soil moisture conditions to determine whether the leak would be expected to be more widespread or more concentrated at the leak point [24].

Potential changes in soil moisture should be always considered to understand how the potential leakage hazard may change over time. One possible approach would be to record the moisture level when a leak was first graded and then to prioritize follow-up surveys in different moisture conditions to verify any change to the leak behavior [23].

The same applies to hydrogen leaks, with adjustments for its physical and chemical properties.

Effect of soil properties on hydrogen leakages

Please note that the data presented are based on numerical simulations focused only on <u>diffusion as the only migration mechanism</u> (without taking into account advection and competition between vertical and horizontal transport).

Despite the shortcomings and limitations, numerical simulations confirm the importance of soil conditions.

Soil texture has a very important impact on hydrogen diffusion.

Hydrogen diffuses more easily in sandy soils, and its diffusion speed, concentration, and range are higher than those in clay soils. The diffusion H_2 coefficient is **six times** higher than that of natural gas. H_2 has a higher diffusion capacity and a more pronounced harmful effect than natural gas [15].

Diffusion height

The maximum height of pure hydrogen leakage diffusion at 60 mins in sandy soil was 3.10 m (1.6 m above the ground), while in clay soil – 2.78 m (1.28 m above the ground) under 1 MPa operation, 1.5 m burial depth, 304 mm pipeline diameter, and 20 mm leakage hole diameter. Under the same initial conditions, the longitudinal diffusion H_2 leakage is significantly higher in sandy soils than in clay soils. Hydrogen diffuses more easily in sand [15].

Another numerical simulation produced significantly higher values. The vertical diffusion height of hydrogen-doped natural gas from sandy soil to the atmosphere after diffusion for 1800 s was 33.8 m and that of loamy soil was 25.41 m, which is a 33% difference between the two [19].

Diffusion range

The hydrogen-blended natural gas (HBNG) diffusion range in sandy, loam and clay soils under the same initial conditions can be defined as large, moderate and small. The farthest danger range (FDR) for 15 % HBR in sandy soil was found to be 2.063 m, in loam – 1.377 m, and in clay – 0.443 m (with increasing HBR, the FDR also increases). Thus, the potential risk of an explosion is highest if the type of soil where the pipeline leak occurs is sandy [16].

Therefore, it is extremely important to have information about soil characteristics: texture (percent of sand, silt, and clay -> Soil Texture Triangle), pore size, porosity, moisture, water saturation, the viscous resistance and inertial resistance coefficients. Clay soils have the highest viscous and inertial resistance, while sandy soils have the lowest diffusion resistance, and, therefore, rapid vertical diffusion of hydrogen is more likely to cause dangerous accidents, while the danger of clay soils is less pronounced.

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