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Reduction of methane emissions from biogas systems and landfills

Methane oxidation treatment for systems with low gas fluxes and low methane concentrations

IEA Bioenergy: Task 37

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Definitions

Autothermic

Technical oxidation process where the necessary heat comes only from the gas to be treated, without addition of heat.

Biomethane

Renewable gas that is produced from biological processes, such as anaerobic digestion in landfills or biogas plants. Typically, landfill gas and biogas are upgraded to remove carbon dioxide and produce a gas with a methane content of at least 96%, which matches natural gas characteristics referred to as biomethane.

Clean Gas

Gas which exits the methane oxidation process and is released into the environment.

Diffuse emission source

For the investigation of emissions at an installation, the definition can be adopted by analogy with the definition in the UN-ECE PRTR Protocol and EC PRTR Regulation No 166/2006/EC: Small or distributed sources whose combined effect may be significant, and for which it is not practicable to measure them individually and obtain a source specific value.

Emission sources

Location of the discharge of the emission (e.g. methane) into the environment. The properties of sources can take many forms. There are captured sources (e.g. piped) and area sources, known and unknown, temporally constant and varying sources.

Fuel gas

Gas which is fed into the combustion or oxidation process and contains the energy carrier (in the context of this study the off-gas of a process (e.g. upgrading process) that is sent to post-treatment.)

Leakage

A defect in a gas-tight system that causes emissions.

Lean gas

Fuel gas with a low energy content

Methane slip

Unused methane contained in the off gas of a technical processes for the production or treatment of gases (e.g. separation of gases or utilization of energy). Methane slip is caused by technology and is not the same as emission sources, since there may still be downstream waste gas treatment processes.

Methane emissions

Discharge of methane into the environment.

Off gas

In this report, it is synonymous with the term exhaust. Depending on its methane concentration and the regulatory limits in place, off gas is either released as an emission to atmosphere, or sent to post-treatment, such as methane oxidation.

Raw gas

Gas which is fed into the methane oxidation process (mixture of fuel gas and air).

Recuperative heat recovery

The media whose heat is to be transferred are spatially separated. Examples are plate heat exchangers or tube heat exchangers.

Regenerative heat recovery

The media whose heat is to be transferred are not separated from each other. Examples are ceramic storage masses through which the gases alternately flow and thus transfer the heat.

1 Introduction

Methane has a large global warming potential and is a significant contributor to the current warming of the atmosphere, accounting for one third of net warming since the Industrial Revolution. To address the importance of its climate impact, an international campaign to reduce global methane emissions was launched at COP 26 in Glasgow, Scotland in 2021. As of January 2025, the Global Methane Pledge has 159 country participants who have agreed to take actions to contribute to a collective effort to reduce global emissions at least 30 percent from 2020 levels by 2030.

Methane is emitted from a wide range of industrial, agricultural and waste management processes. The capture and treatment of emissions are not always technically and, under current conditions, not always economically feasible. A particular challenge is posed by waste gas streams with low methane concentrations, and low and variable volumetric flowrates. The treatment of such waste gas streams becomes expensive and technically difficult if the quantities collected are small. Its low energy content makes energy recovery difficult and oxidation, without extra energy supply, is only possible with complex technology. For biogas systems and landfills, examples of emission sources with low methane contents and low volumetric flowrates include:

- Anaerobic digestion and biogas utilization (biogas plants)
 - Feedstock (including manure) handling and storage
 - Off gases of Combined Heat and Power (CHP) units
 - Off gases from biogas upgrading
 - Open and non-gas tight digestate storage
 - Processes related to biogas plants such as digestate composting, organic waste handling, etc.
- Landfills
 - Landfill gas that is highly diluted with air due to leaks in gas collection or increasing air content in older, closed landfills

Methane oxidation, through thermochemical or biochemical means, can serve as an effective treatment option for some of these emission sources, eliminating the methane as well as volatile organic compounds. For thermochemical solutions, complete methane oxidation requires high temperatures within the incineration process. This temperature should be provided from the gas to be treated, which is also the fuel gas in order to be able to ensure an autothermic process. Therefore, these solutions depend significantly on the energy content of the fuel respective of the gas to be treated and thus the methane concentration of the gas to be treated. Methane can also be used as an energy source by microorganisms who convert it into carbon dioxide and water. Biochemical processes have been used in landfill aftercare to minimize methane escaping into the atmosphere. Here, technical filters, operated in active or passive modes, can facilitate the work of microorganisms to use the oxygen that diffuses into the cover layer to oxidize the methane emissions. Biochemical oxidation is less developed than thermochemical treatment, but appears to offer options when thermochemical solutions reach their technical limits.

This report presents process technologies for methane oxidation, with a focus on processes that can convert waste gases with low methane concentrations emitted at low flow rates. It begins by describing the properties of methane and the climate change impacts that can result from relatively small sources of emissions (Section 2), and then introduces several emission sources at biogas plants and landfills (Section 3). This is followed by a review of the principles of methane oxidation (Section 4), the technical boundary conditions (Section 5) and a description of the technologies and the applications for which they are best suited (Section 6). Energy considerations are briefly covered in Section 7, and a financial analysis of Regenerative Thermal Oxidation (RTO) technology is presented in Section 8.

The report concludes with examples of real-world applications of methane oxidation technologies.

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2 Properties of methane and impacts of methane losses

Methane has long been valued as an energy-rich gas that is used as a source of primary energy and for chemical production. Its key properties are listed in Table 1. Natural gas is a fossil fuel made up mostly of methane with small amounts of natural gas liquids (NGLs), ethane, propane, butane, and pentane. Methane can also be produced from anaerobic biological activity during the decomposition of organic matter in landfills and biogas plants. Here methane typically co-exists in a 55:45 volume percent mixture with carbon dioxide. Increasingly landfill gas (LFG) is being recovered and biogas plants are being built to capture this renewable energy from organic wastes. When the carbon dioxide (CO₂) is removed from these gases in an upgrader, the resulting gas, containing at least 96% methane, is referred to as biomethane.

There are many anthropogenic sources of methane emissions, including emissions from agriculture (livestock production) or energy-related emissions from oil, gas, coal and bioenergy. The waste sector and biomass burning also contribute significantly (IEA 2021). These emissions are generally very low in methane concentration, but collectively these many diffuse sources are increasing the methane concentration of the atmosphere.

Methane is the second largest source of greenhouse gases (GHG) after carbon dioxide. It is relatively short-lived with an average residence time of 12.4 years in the atmosphere, after which time it is converted into CO₂. Although methane is present in much lower concentrations than CO₂, it has a much larger global warming potential (GWP). Over 20 years, methane has 84 times the GWP of CO₂; over 100 years, according to Myhre et al. (2013), it still has 28 times the GWP of CO₂. If the atmosphere continues to warm, it is expected that larger amounts of methane will be released because of the thawing of permafrost. Furthermore, there are large quantities of methane hydrate in the sea and in permafrost, although the effect of progressive climate change on these deposits has not yet been conclusively determined.

For these reasons, attention is being placed on capturing diffuse emission sources and the treatment of emissions with low gas fluxes and low methane concentrations.

Table 1: Methane properties

Methane properties			
Elemental formula	CH ₄	Density at 1013 mbar and 273.15 K	0.7175 kg/m ³
CAS Number	74-82-8	Ignition temperature	595 °C
State of aggregation at 1013 mbar and 293.15 K	Gaseous	Temperature class	1
Colour	Colourless	Lower explosion limit	4.4 vol.%
Odour	Odourless	Upper explosion limit	16.5 vol.%

Methane properties

Molecular weight	16.04 g/mol	Solubility in water at 293.15 K and 1013 kPa	24.4 mg/l
Melting point	-182.47 °C	Max. laminar flame velocity	43 cm *s ⁻¹
Boiling point	-161.52 °C	Flame temperature	1,970 °C
Higher Heating Value	55.5 MJ/kg	Lower Heating Value	50 MJ/kg

2.1 IMPACT OF METHANE LOSSES AT BIOGAS PLANTS

The objective of well-functioning biogas plants is to maximize the conversion of organic matter into biogas with high methane concentrations, and to recover this methane for use as renewable energy. Given that the sale of biogas energy often is the main source of revenue of a biogas plant, a financial imperative exists to maximize methane capture at biogas plants. However, there will be some unplanned losses. Methane losses can be described as structural, operational or accidental (Hurtig et al, 2025) and they can be emitted at different stages of the biogas production chain: e.g., storage and pre-treatment of feedstocks, leaks from the reactor and equipment, biogas utilisation and digestate storage. Emissions cannot totally be avoided but they can be reduced with careful planning and good operation of the digestion plant, including choice of components in the first place, timely maintenance, and good practices when handling and using the end-products - gas and digestate.

Studies from different European countries report great variation on measured methane emissions from biogas plants, from 0.02 to 23.8 vol. % of produced methane (e.g. Reinelt et al 2022; Vergote et al 2020; Fredenslund et al 2018; Reinelt et al 2017; Liebetrau et al 2013). Although the variation can be explained by different reactor and operation types, raw materials and digestate processing methods used, and the size and age of biogas plants, higher methane emissions have been measured at mostly older biogas plants digesting sewage sludge (Scheutz & Fredenslund 2019).

The Finnish KEBIO-project (Luostarinen et al 2023) evaluated the climate impact of potential methane emissions from a biogas plant using the Life Cycle Assessment (LCA) method (ISO 14040). The objective of the project was to estimate how different practices in biogas plants can affect GHG emissions. The emission estimates were based on a literature review and expert evaluations of typical Finnish practices and circumstances. As the intention was to evaluate the potential risks posed by the methane emissions, a range of minimum and maximum values was estimated for the digestion process, pressure control/relief valves, maintenance, and from biogas upgrading to biomethane. In addition, the potential methane emissions from digestate storage were estimated based on the hydraulic retention time (HRT) of the process, i.e. calculated from the amount of degradable material remaining in the digestate.

One of the examples evaluated in the Finnish project was a centralized biogas plant with a total feedstock/substrate capacity of 200,000 wet tonnes per annum. Wet digestion technology was used and the feedstock consisted of pig slurry (40%), poultry manure (15%), separated solid fraction of cattle and pig slurry (15%), grass biomass from nature management fields and green fallows (10%), and side-streams from the food industry (20%).

The maximum and minimum methane emissions (Table 2) are expressed as a percentage of the biomethane production. It was assumed that minimum emissions could be achieved with careful planning and good operation of the plant, while maximum values presented a worst-case scenario. The emissions from open digestate storage were based on the assumptions that maximum methane emissions would be produced if the HRT was 20 days, while minimum emissions would be achieved with an HRT of 50 days. In addition to methane losses, the total emissions from the plant are affected by all the selected practices, so the total emissions are not directly proportional to the factors in Table 2. More detailed descriptions of the comparisons can be found in Luostarinen et al 2023. These are examples for the specific case of Finland, other countries have different situation. In Germany for instance, a minimum retention time of 150 for agricultural plants is required and new digestate storage tanks have to be built gastight covered. On a CO₂-equivalent basis, it was found that the methane emissions were close to three times higher in the maximum scenario (25,980 t CO_{2eq}) than in the minimum scenario (8,150 t CO_{2eq}).

Table 2: Minimum and maximum methane emissions at a centralized biogas plant (200,000 t/a).

	Maximum methane emissions (% CH ₄ prod.)	Minimum methane emissions (% CH ₄ prod.)
Digestion process	0.31	0.1
Pressure control/relief valves	3	0.5
Maintenance	2	0.1
Biogas upgrading	3	0.1
TOTAL	9	0.8
Open digestate storage	8	2

In Europe, GHG emission reductions from improved management of manure can be included in GHG accounting (according to Renewable Energy Directive II (RED II, 2018/2001/EU)). When the emission reductions from improved manure management are included in the Finnish example, the total GHG reductions, compared to fossil fuels, are 123% for the minimum scenario and 52% for the maximum emissions scenario. This indicates that biomethane produced under the maximum scenario would not achieve the 65% GHG reduction required by RED II for the transportation sector. Under the minimum scenario, biomethane production could meet the RED II threshold even without credits for improved manure management.

In this example, the highest emission reduction (compared to fossil fuels) was obtained when the HRT of the process was sufficiently long to ensure the least emissions were released during digestate storage, and when the operational procedures were effective, i.e. including frequent leak detection and minimizing emissions release during maintenance. However, it was also shown that if digestate storage had a gas tight cover with gas collection that resulted in no emissions, the total GHG emissions of the biogas plant in the maximum scenario would decrease by about 40%, over 10,000 t CO_{2eq}.

The high GWP of methane leads even small methane emissions to have a significant impact on the GHG balance of the biomethane produced from the biogas plant. The size of this effect can be explored by examining the methane loss or slip at the biogas upgrader in CO_{2eq} emissions. Within RED II, the eligibility criteria for biomethane require a minimum GHG reduction to be achieved compared to a fossil fuel comparator (FFC) that is valid for the sector of the biofuel utilization. Table 3 presents the maximum CO_{2eq} emissions allowed for biomethane production according to the RED II and the CO_{2eq} emissions that correspond to a given methane slip. For a methane slip of 0.5%, the resulting emissions represent 8% of the allowed emissions for biomethane used in the transport sector. Given that there are other sources of methane emissions in the biogas plant, such as the parasitic energy demand and feedstock supply, that might not be easy to reduce or abate, the methane emissions from upgrading equipment process should be as low as possible.

Table 3: Methane slip in the context of the eligibility criteria of RED II (RED II 2018)

Methane slip	CO _{2eq} emissions per kWh biomethane	CO _{2eq} emissions per MJ biomethane	Eligibility criteria RED II - 65 % reduction FFC	Eligibility criteria RED II - 70 % reduction FFC	Eligibility criteria RED II - 80 % reduction FFC
% of biomethane to grid/fuel	g CO _{2eq} /kWh	g CO _{2eq} /MJ	g CO _{2eq} /MJ (FFC: 94 for transport)	g CO _{2eq} /MJ (FFC: 80 for heat)	g CO _{2eq} /MJ (FFC: 80 for heat)
0.2	3.92	1.1	32.9	24	16
0.5	9.8	2.7	32.9	24	16
1	19.6	5.5	32.9	24	16
1.5	29.4	8.2	32.9	24	16
2	39.2	10.9	32.9	24	16
2.5	49	13.7	32.9	24	16
5	98	27.3	32.9	24	16

The FFC for electricity is 183 g CO₂/MJ, which results in much higher emissions allowed for renewable electricity provision.

The RED II standard values assume that 0.03 MJ CH₄/MJ biomethane are the methane emissions in the off-gases of the upgrading processes: Pressure Swing Adsorption (PSA), Pressure Water Scrubbing (PWS), Membranes, Cryogenic, and Organic Physical Scrubbing (OPS). This represents 3 vol.% of the biomethane, and is likely higher than methane slip from today's biogas upgraders. Nevertheless the emissions are significant and, as explained in the sections that follow, can be reduced using different upgrading technology (e.g. Chemical Scrubbing) or by post-treatment of the off-gas.

3 Methane emissions sources at biogas plants and landfills

Treatment options for methane emission sources depend on the nature of the source and the characteristics of the methane emission requiring treatment. In the case of the biogas plant, the emission sources can be structural, operational or accidental in nature, with the structurally-related emissions arising from the regular operation of a particular equipment. Depending on how easily the emission can be collected, its methane concentration and the size and variability of the gas flow, methane oxidation technology may be suitable for post-treatment. Similarly, there are opportunities for methane oxidation technology to be applied at landfill sites. As they typically exist for decades, landfill emission sources are characterized by declining gas production rates and methane concentrations, and changing gas composition over time. This dynamic feature of landfill gas production influences the treatment strategy and suitability of methane oxidation technologies. Three sources of methane emission are described in further detail.

3.1 OFF GASES FROM BIOGAS UPGRADING

At biogas upgrading plants, there is an off gas stream enriched with CO₂ with methane concentrations in the low single-digit range, on a volume basis. Depending on the type of gas separation technology, the off gas may also contain air, oxygen, hydrogen sulphide or water vapour. Methane losses from biogas upgrading can be reduced in a number of ways, namely:

- 1) Optimization of biogas upgrading technology.
- 2) Combination of biogas upgrading and methane oxidation in post treatment.
- 3) Combination of biogas upgrading and CO₂ liquefaction; if the liquefaction off gas is recycled back to the inlet of the upgrader or utilized, the methane emissions can be reduced.
- 4) Parallel upgrading (e.g. for a local fueling station) and CHP operation; the off gas from the upgrading process can be recycled into the raw gas and used in the CHP. This works only for a fraction of the biogas to the upgrader, i.e. until the methane concentration in the raw gas is too low for efficient upgrading and CHP operation.

The main types of upgrading technologies include water scrubber, pressure swing adsorption (PSA), membrane separation, and chemical scrubber (amine wash). As shown in Figure 1, in Europe, membrane technology has been increasing its market share over the last few years. There are differences among the technologies in terms of the composition of the off gas, and the volumetric flow and concentration of methane in the off gas.

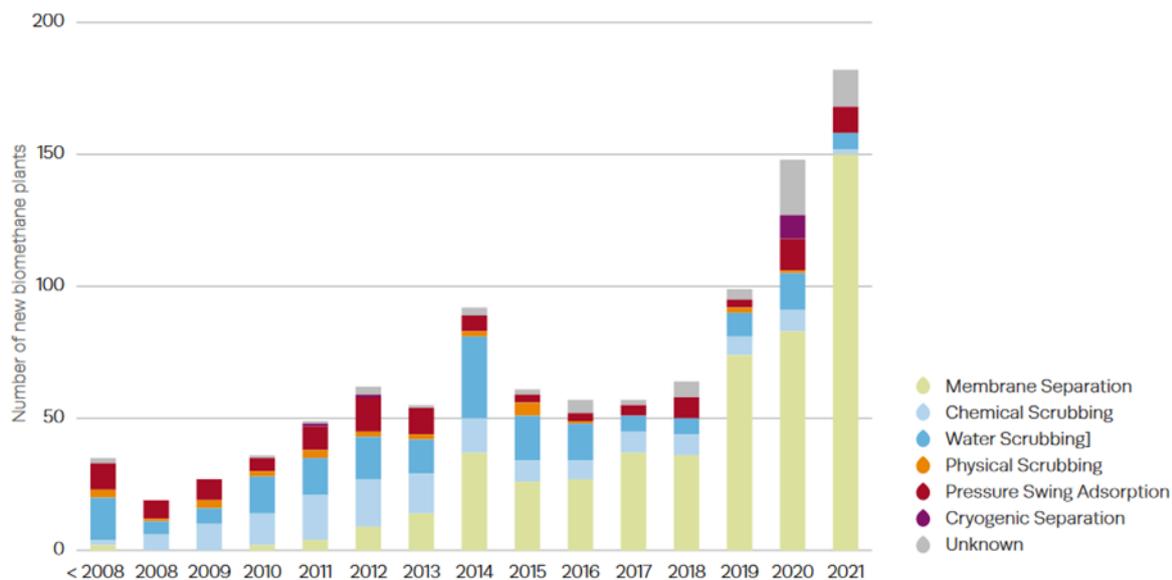


Figure 1: Biogas Upgrading technologies adopted in the EU (EBA 2022)

The methane slips of the different upgrading technologies appear to fall within a similar range. As the biomethane industry has been developing and the technology continues to evolve, the methane slip has been significantly reduced for all technologies. Broadly speaking, the methane slips of PSA and water scrubber units tend to be above 1 %, while for membrane technologies it falls between 0.5 and 1 % and for chemical scrubbing processes (amine wash) it falls below 0.1 % (Wechselberger et al 2023). Membrane separation technology can control the methane slip by means of the number of membranes used, dimensions of the membrane and operation of the unit.

To better appreciate the size (i.e. mass and volume) of the off gas flows,

Table 4 and Table 5 present the methane flowrates and concentrations for a range of biogas upgrading plant sizes treating biogas from 92 to 2,963 m³/h and different methane slip scenarios (0.2, 0.5 and 1%). In addition to the volumetric flow, the methane concentration in the off gas is an important factor for the selection of suitable treatment technologies.

Table 4: Volumetric Flows for different sizes of Biogas Upgrading Plants

	Capacity of Biogas upgrading plant – volumetric flow (m ³ /h)					
Biogas (on a volume basis, composed of 54% CH ₄ and 1% other gases, remainder as CO ₂)	92	185	370	740	1,481	2,963
Methane	50	100	200	400	800	1,600

	Capacity of Biogas upgrading plant – volumetric flow (m ³ /h)					
Carbon dioxide	41	83	167	333	666	1,333
Methane slip within biogas upgrading process	Volumetric flow of methane in the off gas (m ³ /h)					
0.2%	0.1	0.2	0.4	0.8	1.6	3.2
0.5%	0.2	0.5	1.0	2.0	4.0	8.0
1%	0.5	1.0	2.0	4.0	8.0	16.0

Table 5: Resulting methane concentrations in the off-gas of the upgrader

Methane slip	Methane concentration in the off-gas of the upgrader (only CO ₂ considered in off-gas; Biogas of 54% CH ₄ and 1% other gases, remainder as CO ₂)
% of biomethane produced	vol.%
0.2	0.24
0.5	0.6
1.0	1.2

Also, different upgrading technologies produce off gas / waste gases with different compositions. Few studies have been carried out where comparative measurements were made for different technologies. Kvist and Aryal (2019) present data from 9 installations in Table 6. While amine scrubbing and membrane technology produce oxygen-free and nitrogen-free gas, the off-gas from pressurized water scrubbing (similar to polyglycol scrubbing) contains large amounts of air due to the stripping process.

Table 6: Composition of off gas at 9 biogas upgrading plants (Kvist and Aryal (2019))

Gas quality from various points in upgrading units.

SN	Biogas plant	Gas quality (%)		Waste gas (%)	Methane loss (%)
		Before upgrading	After upgrading		
1	Amine based	CH ₄ (45.19), CO ₂ (53.50), N ₂ (0.76), and O ₂ (0.40)	CH ₄ (99.0), CO ₂ (0.50), N ₂ (0.40), and O ₂ (0.10),	CH ₄ (0.06), CO ₂ (99.94), N ₂ (0), and O ₂ (0),	0.07
2	Amine based	CH ₄ (67.03), CO ₂ (32.92), N ₂ (0.01), and O ₂ (0.01)	CH ₄ (98.50), CO ₂ (1.0), N ₂ (0.3), and O ₂ (0.2)	CH ₄ (0.07), CO ₂ (98.92), N ₂ (0), and O ₂ (0),	0.04
3	Amine based	CH ₄ (58.68), CO ₂ (39.42), N ₂ (1.52), and O ₂ (0.37)	CH ₄ (99.06), CO ₂ (0.36), N ₂ (0.58), and O ₂ (0)	CH ₄ (0.09), CO ₂ (99.9), and N ₂ (0)	0.07
4	Water scrubber	CH ₄ (67.03), CO ₂ (32.92), and N ₂ (0.03)	CH ₄ (98.5), CO ₂ (1.0), N ₂ (0.3), and O ₂ (0.2)	CH ₄ (0.42), CO ₂ (15.1), N ₂ (66.64), and O ₂ (18.16)	1.3
5	Water scrubber	CH ₄ (60.83), CO ₂ (38.75), N ₂ (0), and O ₂ (0)	CH ₄ (98.5), CO ₂ (0.75), N ₂ (0.5), and O ₂ (0.25)	CH ₄ (0.41), CO ₂ (13.1), N ₂ (68.33), and O ₂ (18.16)	1.97
6	Water scrubber	CH ₄ (62.6), CO ₂ (36.8), N ₂ (0.43), and O ₂ (0.1)	CH ₄ (98.17), CO ₂ (0.73), N ₂ (0.78), and O ₂ (0.32)	CH ₄ (0.65), CO ₂ (22.29), N ₂ (60.62), and O ₂ (16.18)	1.7
7	Water scrubber	CH ₄ (62.62), CO ₂ (36.8), N ₂ (0.4), and O ₂ (0.1)	CH ₄ (98.17), CO ₂ (0.73), N ₂ (0.77), and O ₂ (0.32)	CH ₄ (0.28), CO ₂ (14.32), N ₂ (66.98), and O ₂ (17.93)	1.1
8	Membrane	CH ₄ (58.22), CO ₂ (40.45), N ₂ (0.77), and O ₂ (0.54)	CH ₄ (99.0), CO ₂ (0.5), N ₂ (0.4), and O ₂ (0.1)	CH ₄ (0.68), CO ₂ (99.31), N ₂ (0), and O ₂ (0.0)	0.48
9	Membrane	CH ₄ (53.12), CO ₂ (45.86), N ₂ (0.63), and O ₂ (0.36)	CH ₄ (99.0), CO ₂ (0.5), N ₂ (0.4), and O ₂ (0.1)	CH ₄ (0.64), CO ₂ (99.35), N ₂ (0), and O ₂ (0)	0.56

The off-gas composition influences post-treatment to reduce methane emissions and CO₂ recovery. For subsequent methane oxidation treatment, the presence of oxygen in the off gas is advantageous, since the oxygen required for oxidation might not be need to be added.

However, if the CO₂ is to be separated from the off-gas stream and utilised for food grade uses, for example, the air content of the off-gas increases the effort required for CO₂ processing.

The type of upgrading technology clearly influences the need for post-treatment, the size and variability of the off gas flows and the type technologies that might be best suited to reduce the methane concentrations. Some of the considerations that should be taken into account include:

- Pressure swing adsorption (PSA) results in fluctuations in pressure, flow rates and methane concentrations in the off-gas stream. Post-treatment processes would therefore need to be preceded by an equalization tank or intermediate storage tank. Also, here methane slippage typically increases over time, as the molecular sieves are subject to aging and the separation quality declines.
- Membranes have slightly different separation performance dependent on manufacturer and also exhibit ageing effects over time. Process parameters as pressure, recycling rates, number of processing steps and cooling have an impact on the methane slip.
- Water scrubbing has oxygen and nitrogen in the off gas, as well as water vapour. The used water must be replaced from time to time, which can cause fluctuations in the off gas quality. Also, if hydrogen sulphide is present in the off gas, corrosion of the downstream oxidation process may result.
- Very low concentrations of methane would likely prevent the autothermic operation of a methane oxidation technology. Data presented in Table 6 show that the amine wash technology has a very high selectivity, with resulting methane concentrations 8 to 10 times lower than the off gas from membrane systems. Post-treatment might not be required, but if this is not the case, the remaining methane concentrations would not likely be sufficient for autothermic operation.

Methane oxidation technologies can be used to address the low concentrations of methane slip from upgrading systems, i.e. by converting the remaining methane into carbon dioxide. The technologies commonly used in practice for the oxidation of these off gas streams are Regenerative Thermal Oxidation (RTO) and the E Flox process, the latter being used in the past mainly for processes with relatively high slip. They are further described in Section 6.

Efforts by the manufacturers of upgrading technologies to increase methane recovery and reduce methane slip are resulting in lower methane concentrations in the off gas and less thermal capacity. This makes post-treatment increasingly difficult as the technical, thermodynamic and economic limits of a treatment technology are reached. For example, below a certain methane concentration there will not be enough energy for the treatment to be autothermic, and either energy would have to be added or other treatment options would need to be explored. Therefore, the continuous improvements in biogas upgrading technology need to be considered when selecting the appropriate treatment technology.

3.2 Methane emissions from manure storage

The storage of organic materials that contain sufficient water can lead to the formation of methane. Examples of sources of such methane emissions include the uncovered storage of sewage sludge, liquid manure and digestate. The emissions depend on numerous factors including the methane formation potential and the degradation properties of the stored substances, the availability of methanogenic microorganisms, the storage temperature, pH, water content and the storage period.

An estimation of the methane load as a design parameter for temporary storage of organic substances such as liquid manure must consider the kinetics of gas formation, i.e. non-linear dynamics usually modelled as first order kinetics. However, methane emissions are reported in national GHG inventories as % of biogas formation potential, thereby representing an average emission value over the entire time and all manure quantities.

In practice, manure slurry is stored and applied over the course of the year, resulting in an average storage period and the slurry remaining in storage for different lengths of time.

As shown in Figure 2, the non-linear gas formation is adapted to the emission potential of the slurry in such a way that the value specified by the emission inventory is reached on average over the storage period. The emission value assumed for this calculation, $1.84 \text{ m}^3 \text{ CH}_4/\text{m}^3 \text{ slurry} \cdot \text{a}$ was taken from KTBL 2021.

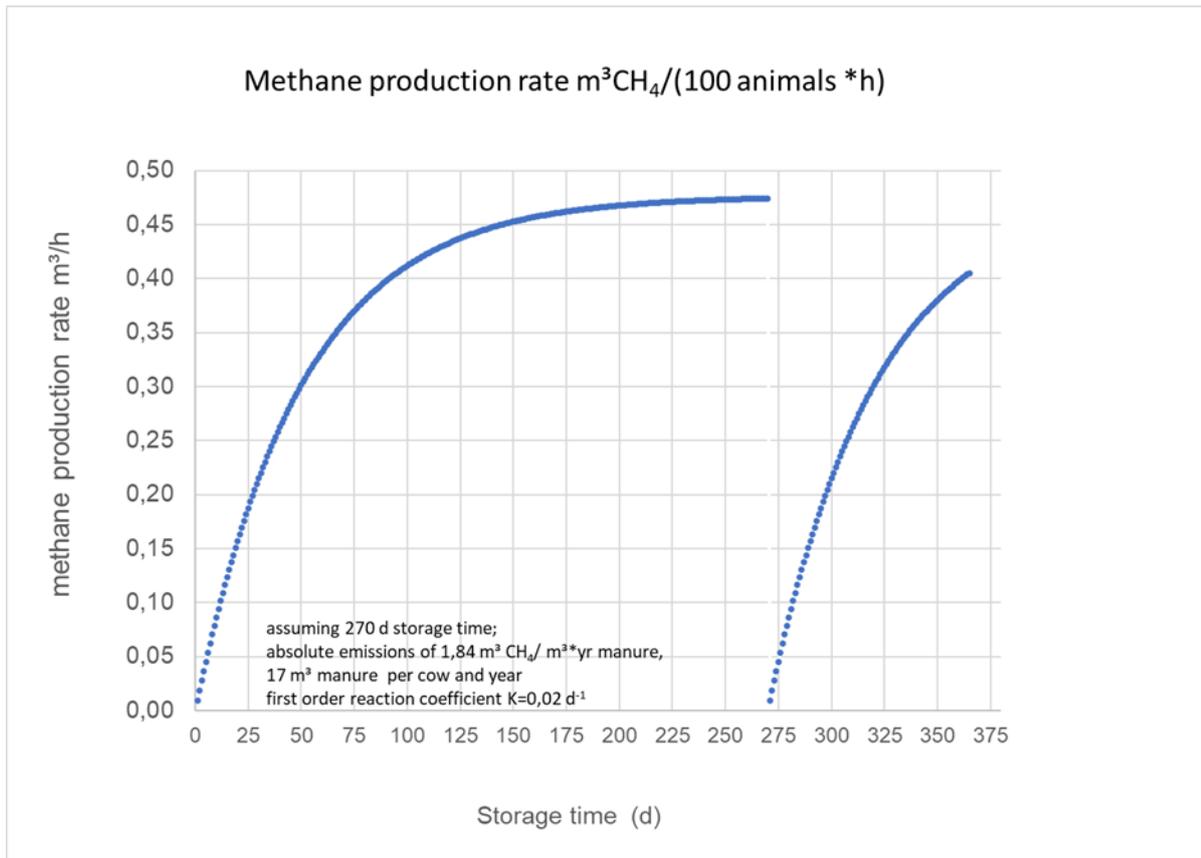


Figure 2: Methane formation rate from a storage tank with capacity for 270 days storage (100 cows)

Depending on how the emissions are captured, the collected gases can be treated in various ways. Due to the gradual filling and abrupt emptying of such storage tanks, the corresponding volumes in the gas space must be balanced. In the case of gas tight covered storage tanks the best solution would be to use gas from other storage units. If gas for balancing is not available, air might be used to balance the changing filling levels. However, by adding air, explosive mixtures of methane and air may be formed and need to be managed. If the cover is not gas-tight, air is constantly entering the headspace, avoiding the formation of explosive mixtures. However, this process needs to be controlled. In the case of continuous extraction of gas from the headspace of a non-gas tight cover, the resulting methane concentration depends on the degree of dilution of produced methane with air.

Oxidation by means of combustion technologies usually has high specific costs since the effort to collect and combust the gas does not match the revenue that can be obtained for the gas or energy. Also, the dynamics of gas production from storage tanks make it difficult to operate. Biological methane oxidation may present an alternative for such applications.

3.3 METHANE EMISSIONS FROM LANDFILLS

Landfill regulations and operations differ from one country to another. Active landfills with ongoing deposition of organic fractions of waste can produce a landfill gas (LFG) with high methane concentrations. However, LFG production and emissions behaviour of old, closed municipal waste landfills follow a different pattern. As gas production decreases due to the depletion of organic material, there is increasing intrusion of air, diluting the LFG and lowering methane concentrations. That is the nitrogen and carbon dioxide concentrations increase, while oxygen is consumed in the landfill.

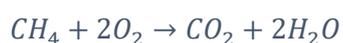
Thus, sites with LFG collection systems are characterized by a decrease in methane load, as well as a decreasing concentration of methane in the extracted gas over time. The methane concentration in the extracted gas depends on the efficiency of the gas collection, the landfill cover and the operation of the extraction system. With sinking LFG production, the amount of gas extracted from the landfill needs to exceed the produced gas in order to achieve high coverage and reduce emissions from the site. An important parameter for ensuring emission reduction is therefore the factor by which the extracted gas quantity exceeds the quantity of produced landfill gas.

If LFG has to be captured and treated, the emissions treatment technology has to be able to adjust to the changing characteristics. For installed methane oxidation technology, this means that a wide operational range would be required in terms of thermal output (corresponding to the methane load captured) and methane concentration in the landfill gas in order to enable a long period of technology utilization. In the aftercare phase of landfill operation, methane concentrations may become too low to be treated technically or economically by thermochemical oxidation technologies, and biological methane oxidation systems may be an option.

4 Principles of methane oxidation

The technical oxidation of methane is mainly used to provide energy, although oxidation is becoming an increasingly relevant means for treating off gases.

Oxidation follows the equation:



Equation 1: Methane oxidation

The reaction is strongly exothermic and the enthalpy of reaction is - 890.95 kJ/mol ($\Delta G_0 = 780$ kJ/mol (Scheutz 2009)). Due to its energy content, methane is a central component of energy supply in the industrial and private sectors in the form of natural gas, biogas, mine gas and landfill gas. A relatively high activation energy is required for the oxidation of methane and thus complete oxidation is only achieved at relatively high temperatures.

Besides the energy content of the fuel gas, the amount of air added plays an important role for the temperatures in the combustion process. Air supplies the combustion process with the necessary oxygen and is a component to control the temperature in the combustion process. Since the heat generated in the combustion process is mainly transferred to the fuel gas/air mixture, a decreasing temperature in the combustion gases can be observed as the methane content in the fuel gas/air mixture decreases. Accordingly, at high methane concentrations in the fuel gas, air is added in excess to achieve cooling and avoid excessively high temperatures during combustion. At low methane contents in the fuel gas on the other hand, the energy is no longer sufficient to heat the gas mixture to the minimum temperature required for complete oxidation. In this case, measures must be taken to reduce heat losses and increase temperature of the raw gas by reuse of heat in order to reach the necessary temperature in the process.

The incineration technology that currently has the lowest heat losses and can therefore operate autothermally (that is, without input of external energy to raise temperatures without auxiliary firing by means of additional fuel injection in normal operation) with the lowest methane content in the fuel gas is Regenerative Thermal Oxidation (RTO) technology.

As an alternative to ensuring high temperatures, the activation energy can be reduced. This means that the required temperature for oxidation can be reduced. This can be done by using catalysts, with catalytic processes for methane oxidation needing a temperature of at least 400-500°C (Bindig et al 2016), compared to combustion processes which require 850-1000°C. In practice, however, the application of catalytic methane oxidation proves difficult, as methane is comparatively stable, oxidation still requires relatively high temperatures, and many "biogases" contain substances such as H₂S that can rapidly deactivate the catalysts.

Besides combustion processes, biological methane oxidation can also take place at moderate temperatures. Methane can be used by microorganisms as an energy source. Methane-oxidizing microorganisms, methanotrophs, are present in soils and water. New technologies to reduce methane emissions from old landfills rely on their ability to oxidize methane. It might be also an alternative for other applications, such as storage tank emissions, but this needs further development.

4.1 THERMOCHEMICAL PROCESSES

The stoichiometry of methane oxidation shows that for 1 molecule of methane, 2 molecules of oxygen are needed (Equation 1). This results in the following (minimum) combustion air requirement for complete oxidation of a given raw gas containing no oxygen, in relation to the methane concentration in the fuel gas.

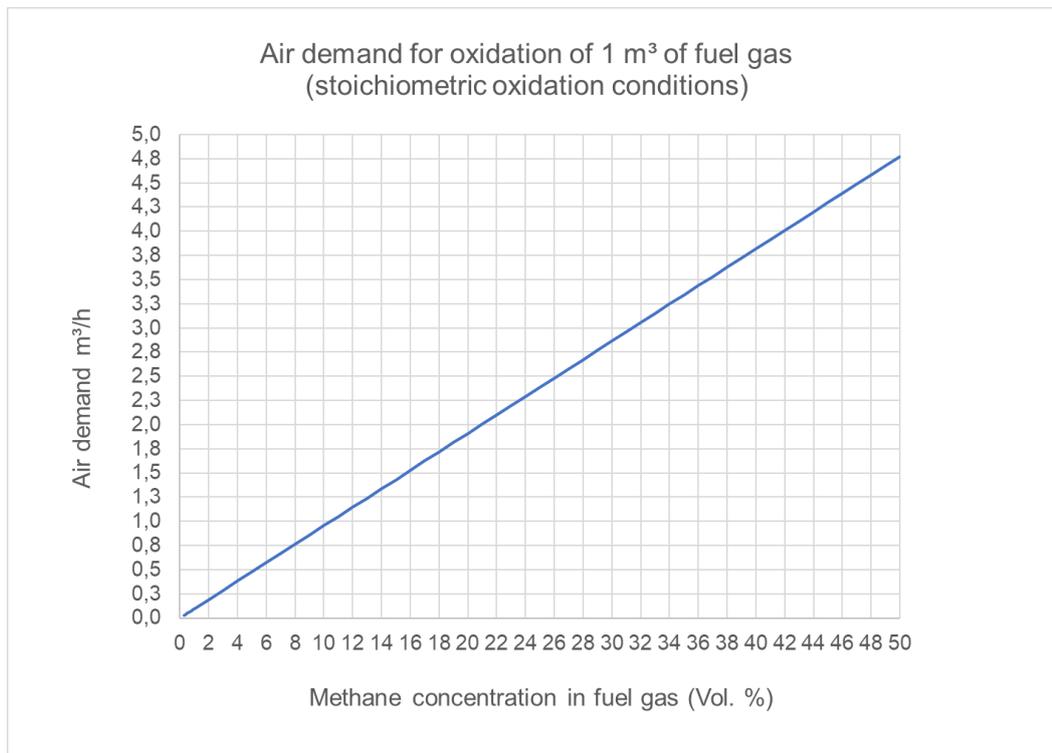


Figure 3: Stoichiometric air requirement as a function of methane concentration

Depending on the desired combustion conditions (temperature and mixing conditions), air is also added over stoichiometric conditions.

If a fuel gas containing 100% methane is burned in a stoichiometric ratio with air (under atmospheric pressure), then, theoretically, the resulting temperature reaches approximately 2000 °C. The temperature can be controlled by the addition of excess air and by the design of the combustion chamber. Operating with excess air cools the process and also optimises the contact of oxygen and methane necessary for oxidation. In the case of high calorific gases, a temperature reduction in combustion is usually desired (in order to protect the material, ensure high oxidation rates and prevent the formation of undesirable products such as NO_x), and this is usually achieved through operation under excess air conditions.

In addition to the amount of air mixed in, the composition of the fuel gas also has an influence on the temperature. As the methane content in the combustion gas decreases, the proportion of inert gas components increases and thus combustion temperatures decrease. In the case of low calorific gases, the high dilution of the fuel gas can lead to low combustion temperatures. If the methane concentration - and thus the energy content of the fuel gas - is low to the point where the required (or desired) temperature increase of the fuel gas/air mixture for complete oxidation can no longer be achieved, the following measures can be taken to improve oxidation:

- Reduce heat losses in the process
(Reduction of radiation losses of the combustion device)
- Preheat the incoming gases with recovered off gas heat (recuperative or regenerative)
- Add high calorific gases to the fuel gas or electrical heating of the heat exchangers

The resulting degree of burnout is influenced by the technology of combustion, the dimensioning of the combustion chamber and the operation. The following parameters impact the oxidation:

- Retention time in the combustion process,
- Oxygen,
- Temperature and temperature distribution, and
- Turbulence of the gases.

The national regulations for the combustion process use these factors in order to ensure nearly complete combustion by specifying technical parameters. For landfill gas flared continuously, German guideline TA Luft defines conditions of minimum 1000 °C at 0.3 s retention time in the combustion chamber. For flares that are only used temporarily as secondary gas utilization (back up of emergency flare), 850 °C is prescribed. RTOs, which have a significantly higher retention time, can also achieve a high degree of burnout at lower temperatures. In Germany there are no temperature regulations here; the efficiency of the burnout is regulated by the limits of methane in the off gas (20 mg C*(m³)⁻¹).

4.1.1 Oxidation or combustion with flame

In combustion with flame formation, the fuel gas is either premixed and ignited in the combustion chamber or mixed with air directly in the combustion chamber and ignited. If the energy content of the fuel gas is high enough, air can be mixed in without specific measures of flow control. The combustion then forms a flame which can be monitored via UV sensors. With decreasing methane concentration in the fuel gas (below 25 vol.%), the air supply needs to be more precisely controlled.

4.1.2 Flameless oxidation

Flameless oxidation takes place by oxidation of combustible gases without the formation of a flame or flame front. This is achieved by mixing fuel gas and air and increasing the temperature of the fuel/air mixture according to the low methane content fuel gas. The resulting oxidation occurs at a more uniform temperature and occupies a larger volume than that of a flame. Due to the lower energy density in combustion, the formation of a flame is avoided. Flameless oxidation allows complete and clean combustion even without high excess of oxygen. The formation of NO_x is largely avoided by the uniform temperatures (Energie-Lexikon 2024).

Flameless operation is achieved, for example, by RTO technology. Furthermore, combustion devices that operate with recuperative heat recovery can realize complete oxidation without a detectable flame. These include flox burners or flare-like combustion systems with external preheating of air and fuel gas by means of heat exchangers. Gases with low energy content must be preheated, which, in the case of some technologies, is done outside the combustion chamber. Due to the ignition temperature of methane of approximately 600 °C, explosion protection measures must be followed.

4.1.3 Catalytic treatment

Catalysts can reduce the activation energy of chemical reactions. If catalysts are used in post-combustion, hydrocarbons can be oxidized at significantly lower temperatures compared to combustion processes. As methane is relatively stable, effective catalyst materials such as platinum are expensive and subject to rapid inactivation when used in biogas applications due certain gas constituents that diminish the effectiveness of the catalyst. For these reasons, the use of catalysts has not yet been established in the biogas and landfill sectors.

4.2 MICROBIOLOGICAL PROCESSES

Methane is an energy-rich gas and thus also suitable as an energy source for microorganisms. Microbiological processes are bound to basic physical, chemical and thermodynamic principles and essential process properties can be derived from the consideration of resulting parameters such as solubility of gases, mass transfer rates, pH or temperature. For the use of gaseous substrates, it must be noted that these must be present in the water phase in order to be taken up by the microorganisms. The mass transfer of the substrate into the cell takes place diffusively following the concentration gradient. Methane and oxygen are the base elements for methane oxidation and are much less soluble in water than carbon dioxide, ammonia or hydrogen sulphide (Stegmann et al. 2003).

In addition, the solubility of the gases also depends on the temperature, with lower temperatures leading to greater solubility. Consequently, the substrate concentration available to the microorganisms is greater when the water temperature is lower. However, biochemical processes are less effective at low temperatures.

Since the growth of microorganisms in certain concentration ranges is dependent on the concentration of the substrate (e.g. according to Michaelis Menten kinetics), one of the limiting variables is the availability of substrate. According to Stegmann et al. 2003, the majority of methane-oxidizing microorganisms in culture are mesophilic with respect to temperature and classified as neutrophilic with respect to pH, although methane-oxidizing organisms have also been isolated in extreme environments. The majority of methane-oxidizing microorganisms depend on methane, methanol or formaldehyde as a substrate, and are therefore also referred to as methylotrophs (Scheutz et al. 2009).

In addition to the supply of methane and oxygen, methane oxidation performance is influenced by a variety of environmental conditions. These include soil structure, temperature, water content in the filter material and available nutrients such as nitrogen and trace elements.

Temperature has a considerable influence on methane oxidation performance by microorganisms. However, activity can still be observed at low temperatures, even if it is significantly reduced. In Scheutz et al 2009, 5-10 °C is mentioned as the range in which methane oxidation significantly declines. Moisture is needed for microbial processes, but too much moisture can restrict gas movement and the diffusion of substrates. The pore volume available for gas exchange is another important parameter in methane oxidation. Methane-oxidizing microorganisms are obligate aerobes and are still active even at oxygen concentrations as low as 1 to 3 vol.%.

For each mole of methane assimilated, methane oxidizers require 0.25 ml of N (Scheutz et al. 2009). Type II methane oxidizers, however, can take up nitrogen from the air, but this is energetically disadvantageous compared to the uptake of ammonia. Limitations due to nitrogen deficiency have already been demonstrated, but the mechanisms are not yet well understood.

The occurrence of exopolymeric substances (EPS) in methane oxidation experiments has been often referred to in the published literature. EPS formation leads to blocking of the soil pores, which in turn results in a decrease in performance since methane escapes through preferential flow paths. In passive systems the formation of EPS was not observed, and intermittent feeding may have an influence on the formation of EPS. The production of intercellular polysaccharides is a strong indicator of methane overabundance (Scheutz et al. 2009). Good control of pore volume available for gas migration helps lower EPS formation.

The optimal pH value for methane oxidizers is between 5.5 and 8.5 (Scheutz et al. 2009). The buffer capacity of the filter material has a great influence on the stability of the pH value. Metabolic products of methane oxidation normally decrease the pH value. Substances such as difluoromethane, dichloromethane, methyl fluoride, acetylene, ethylene and ammonium were found to inhibit methane oxidation. Pesticides also have a negative impact on methane oxidation rate.

Data on process parameters for actively ventilated methane oxidation filters are not available for industrial scale applications.

To ensure sufficient substrate supply in the liquid phase, high concentrations of methane and oxygen in the gas phase and sufficient residence time in the system must be guaranteed.

The structure of the filter material should allow a good transition of the substrates from the gas phase to the water phase. Due to the possible occurrence of EPS formation, the filter should be monitored and "over-supply" should be avoided. pH value should be in the neutral range. Controlling the temperature of the gas mixture or in the filter is rarely possible in practical applications. Methane oxidation releases heat which can lead to particular active zones with higher temperatures. In any event, the temperature in the filters should not be below 10°C.

As noted in the published literature, very different methane oxidation efficiencies have been achieved, whereby due to the large number of relevant and interacting parameters such as methane concentration in the supplied mixed gas, dwell time in the filter, filter material, filter structure, oxygen concentration, humidity, inoculation material, and temperature, the results are very difficult to compare or to transfer into practice. However, it can be stated that the following parameters play a major role:

- temperature,
- methane load (and the associated residence time within a biosystem),
- availability of interconnected air-filled pores allowing gas migration,
- the presence of molecular oxygen,
- humidity resp. availability of water.

5 Technical boundary conditions for methane oxidation processes

5.1 PROPERTIES OF THE RAW GAS

For the technical oxidation of methane, the conditions of the upstream process are important as they affect both the volume flows and quality of the gas. The quality parameters of particular interest include:

- methane concentration,
- oxygen concentration,
- types and concentrations of contaminants and/or
- concentrations and composition of other gases (e.g. water vapour, hydrogen sulphide)

For the design of technical methane oxidation systems, the temporal variations of the gas composition and gas quantities are also important design parameters. Table 7 shows how these parameters can vary with the off gas to be treated.

Table 7: Composition and quantities of gases for treatment

Application	Methane concentration in the gas to be treated	Volume flow methane	Oxygen content	Contaminants
	vol. %	m ³ /h	vol. %	
Biogas upgrading off gas treatment	0.1-2	Depending on throughput and technology of plant Example: 200 m ³ /h off-gas with 1.2 % vol. CH ₄ : 2.4 m ³ /h	Depending on technology: 0-18	Depending on upstream treatment, possibly H ₂ S, siloxanes
CHP off gas treatment (Example: 800 kW _{el} ; lambda 1.5)	around 0.2; in some cases, significantly higher-	Off gas: 1,800 ppm CH ₄ 3.5 m ³ CH ₄ /h Overall gas: 1,940 m ³ _n /h	at λ 1: 0 at λ 1.5: 6.2	Sulphur oxides, hydrogen sulphide, formaldehyde, NO _x
Off air Slurry storage	0 - 2	0 - 5 m ³ /h (max.:1,000 cattle)	18 - 21	Sulphur compounds Ammonia
Landfill gas	0 - 65	Depending on the size of the landfill, age of waste and filling methodology	0 - 21	Siloxanes, possibly chlorine and fluoride containing compounds, sulphur compounds

Industrial processes such as biogas upgrading are designed for continuous operation and thus generally show low fluctuations in terms of gas quality and quantity during post-treatment of waste gases. In other applications, such as treatment of landfill gas, the composition of the gas and the delivered firing thermal output are subject to long-term changes, which should remain within the limits of the technology for as long as possible, in order to achieve a high degree of return on the investment. Here, the largest possible capacity range in which the plant can be operated is desirable.

In the short term, fluctuations in gas flows and composition can be observed in several applications and need to be balanced by the technology. Also, in the conceivable treatment of off gas air from manure slurry storage or sewage sludge storage, very different gas flows and compositions are produced with filling and emptying of storage tanks.

5.2 ENERGY RECOVERY FROM OFF GASES

Regulatory requirements are decisive for the applicability and design of methane oxidation technologies. In addition to specifications regarding maximum permissible concentrations, specifications regarding temperatures to be achieved and residence times in the combustion process can also influence the suitability and dimensioning of the technologies.

The process should be implemented as efficiently as possible in terms of energy and costs. In addition to the use of surplus energy, the energy requirement of the treatment technology (parasitic energy demand) should be as low as possible.

When using gases with sufficient methane content, energy in the form of heat and electricity can be easily extracted from the combustion process and used (e.g. CHP). Below methane concentrations of approximately 20-25 vol.%, the available technologies cannot use the gas within CHP technology. Currently, below 20-25 vol.%, flares or flameless oxidation are used and then only heat can be extracted.

The lower the methane content in the raw gas, the lower the clean gas temperature will be and the more difficult it is to extract heat energy at a usable level.

Additionally, the energy demand to be covered is often not constant (particularly in the case of heat supply e.g. for housing). Since methane oxidation is more effective in regards to emission reduction than energy substitution, the provision of energy is usually of secondary importance.

5.3 EMISSION LIMITS FOR TREATMENT TECHNOLOGIES

Further specifications come from the regulatory authorities regarding permissible emissions. It should be noted here that there are not always specific limits for methane.

In Germany, for combustion engine plants, a methane threshold was introduced in 2022, setting the limit to 1300 mg/m³ in the off gas (see Table 7: 1300 mg equals 1800 ppm, assuming a lambda of 1.5 this translates to a slip of 1.76 % of the utilized methane)

Depending on the national or local regulations, the off-gas from upgraders can either be emitted or needs to be further treated. The regulations usually refer to clean air regulations as well as regulations associated with the eligibility for renewable gas support mechanism. For upgrading facilities in Germany, there are several applicable laws including the subsidy scheme, which requires, within the Gas Grid Access Ordinance, a maximum permissible methane emission from the processing plant to be 0.2 vol. % of methane fed into the grid. This value is reliably complied with when using amine scrubbing; however, all other upgrading processes have a higher methane slip and require post-treatment of the off gases from the upgrader.

Other countries have different limits for methane emissions from upgrading, many of them have just been changed or are under discussion. Denmark for instance allows a methane slip of 1 %; France, according to plant size, between 0.5 and 1 %; UK has no limit at present; and Austria recommends a 0.2% limit in its technical guidelines.

As far as emissions in general are concerned, the requirements of the TA Luft in Germany (Technical Instructions on Air Quality Control) apply to operating permits as well. Organic Substances are limited to 0.5 kg/h as mass flow or the mass concentration of 50 mg/m³ apply to off gases in general. Specifically, for afterburning facilities (applicable for RTO) a threshold of 20 mg/m³ is required. For flares at landfills, a temperature of 1000 °C at 0.3 s residence time is defined. The requirements are not entirely consistent, and different technologies can have different requirements for incineration of the same gas.

Another important aspect relates to the use of the biomethane. In Europe, the RED II requirements provide standard values for the certification of renewable fuel, including for the emissions from technologies.

These standard values distinguish between biomethane produced with and without post-treatment of off gases from upgrading. Due to its low methane slip, amine scrubbing is treated like a plant with post-treatment. Plants without post-treatment have to use quite high values for their emissions (0.03 MJ CH₄/MJ biomethane (RED II 2018)), however this value can be substituted by an actual emission value that is supported through measurement. In Germany, the value of biomethane in the transport sector depends on the GHG reduction potential of the certified gas (biomethane). Accordingly, low methane emissions can directly result in higher selling prices for the biomethane. Therefore, the minimization of the emissions has economic motivation.

At landfills, emissions are minimised by extraction and treatment of landfill gases. At older, closed sites the thermochemical oxidation processes might no longer be possible due to poor gas quality and low methane flows. Further methane oxidation is ensured by directing the methane load into passive or active microbial oxidation systems. There are examples for regulations for biological systems according to which they must be constructed and the concentrations measured at the surface need to comply with landfill regulations (e.g. Laga 2020). In general, a model for the remuneration of avoided GHG emissions would be desirable. In this way, an incentive could be created in applications such as landfills or slurry storage to invest in technology to reduce emissions. Small emission sources are of particular interest here, since they are large in number, but the capacity is usually so small that specific treatment costs are high. Any remuneration system requires a quantification of loadings and emission reduction. This might pose a severe challenge for some systems, such as landfill covers or methane oxidation windows.

5.4 OPERATION OF THE TREATMENT TECHNOLOGY

Methane oxidation plants are used as primary or secondary combustion facilities. Secondary combustion facilities serve as a backup and only operate a few hours per year. For this reason, these plants are subject to other legal requirements in certain jurisdictions (such as Germany). Within the scope of this study, only technologies intended for continuous operation are considered.

5.4.1 Operation - start/stop

Planned and unplanned interruptions do occur during operation of technical systems. Due to technical or safety-related specifications, shutdown and restarting is associated with a certain time and technical effort. Since low methane concentrations mixed with air may lead to explosive mixtures, it is necessary to check whether an explosive mixture is present before (re)starting.

In the case of technologies that require external sources of heat for start-up, such as preheating the gases and air, the start-up process is associated with a heating-up process after a longer period off line. This entails certain limitations in the rate of temperature rise resulting from the material properties of the heat exchangers; in addition, the rate of start-up depends on the capacity of the burners, or on the heating capacity of the electric heater. The shutdown of the plant is again limited by a cooling rate; for example, maintenance work on the heat exchangers requires that they be cooled down.

Systems with preheating, such as RTOs, require time to cool down after shutdown and to preheat after start up of operation. Therefore, short-term shutdowns of these systems can be tolerated, since the plant does not cool down quickly on its own. Longer downtimes, on the other hand, require time-consuming and energy-intensive heating and should therefore be avoided.

5.4.2 Fluctuations in gas composition or gas volume

Fluctuations in the feed to the plant, whether due to changes in concentration or volume, result in a change in thermal capacity and a change in the throughput of the plant.

Whether the plant can handle this change depends not only on its amplitude of the change but also on the speed of the change. In the case of landfills, for example, the formation of gas slowly declines over a period of years; in the case of older landfills, a strong increase in atmospheric pressure can lead to strong short-term fluctuations in gas quality as a result of intrusion of air into the landfill.

Operation in the event of long-term fluctuations can only be guaranteed by appropriate dimensioning of the periphery and the plant components. Such permanent changes in flow and capacity of the plants can only be realized within certain limits. Typical values for the capacity spread are 1:8 with flares and RTOs, but depending on the manufacturer, these values can also deviate.

Short-term changes are compensated for by the operating regime and the control systems, whereby the above-mentioned limitations with regard to capacity and flow also apply here. If high rates of change are to be expected, the system must be adapted in such a way that the process control can compensate for the changes that occur in such a way that the operating parameters of the plant are maintained. Otherwise, gas with an unfavourable composition may briefly enter the combustion chamber and cause overheating, for example. One example of such short-term changes in the off gas is appearing when treating the waste gases from the pressure swing adsorption upgrading process. Here, short-term peaks in the methane concentration occur during the purging of the columns. These peaks are balanced out via buffer storage and fed to the afterburning system.

In principle, an increase in the concentration of methane in the raw gas leads to a higher clean (or off) gas temperature from the RTO and thus to a greater amount of energy that can be extracted. Since some treatment processes (e.g. membrane upgrading process) can control the methane slip and therefore concentration in the off gas by control of process parameters, a targeted increase of the methane concentration would be conceivable in case of an increased heat demand (e.g. in winter).

6 Technical applications for methane oxidation

In principle, methane oxidation should take precedence over energy provision due to the high climate impact of methane. When converting 1 m³ of methane into electricity and assuming the substitution of an electricity mix of 366 g/kWh (Germany 2020) (see Table 8), this amounts to approximately 1,500 g CO₂ avoided (without considering heat use, GWP of methane 28). If 1 m³ of natural gas with 260 g CO₂/kWh (Fossil fuel comparator RED II) is substituted, then approximately 2,600 g CO₂ are avoided. In contrast, 1 m³ of methane emitted directly without treatment has a greenhouse gas effect of about 19,600 g CO_{2eq} due to the GWP value of 28. It should be mentioned that 28 refers to 100-year time horizon. The GWP of methane is 86 on a 20-year time horizon, making the short-term impact much higher.

Table 8: Emission reduction resulting from different utilization/avoidance scenarios

Volume Methane	Energy/emission	Avoided CO ₂ emissions
1m ³ CH ₄	10 kWh _{thermal}	2,600 g CO ₂ avoidance through natural gas substitution
1m ³ CH ₄	4 kWh _{electric}	1,460 g CO ₂ avoidance through electricity substitution, assuming a grid average of 366 g/kWh and 40 % conversion efficiency of fuel gas energy to electricity
1m ³ CH ₄	0.7 kg * 28 GWP 0.7 kg * 86 GWP	19,600 g CO _{2eq} avoidance through emissions prevention (100 yr time horizon) 60,200 g CO _{2eq} avoidance through emissions prevention (20 yr time horizon)

Accordingly, from the point of view of climate effectiveness, avoiding methane emissions should have priority. Second, emission reduction should have priority over energy recovery in the treatment of methane-containing gases. In best cases the benefits of emission reduction and energy recovery are additive.

For the energetic use of methane-containing gases, CHP (electricity and heat), upgrading to biomethane (all natural gas utilization pathways), boilers and flares (heat utilization) are technologies readily available. Micro gas turbines, fuel cells and Stirling engines can also be used, but so far have seen less adoption. The mere oxidation of methane-containing gases is realised by flaring (possibly with preheating of fuel gas and/or air), the e-flox process, regenerative thermal oxidation (RTO) or catalytic processes.

In the case of biogas and in particular landfill gas, conditioning or gas purification plays an important role. Potential contamination of the raw gas by, for example, hydrogen sulphide, siloxanes and/or dilution with nitrogen needs to be properly addressed.

In general, the lower the energy content of the gas, the lower the off gas temperature and thus the more difficult it is to provide energy or to extract thermal energy. Figure 4 shows the relation between methane content in the fuel gas, air mixing and partition of energy for temperature increases coming from the methane oxidation and the recuperation (regeneration) of energy in the process. (Assumed air addition for $\Lambda = 1,5$, no losses in the process).

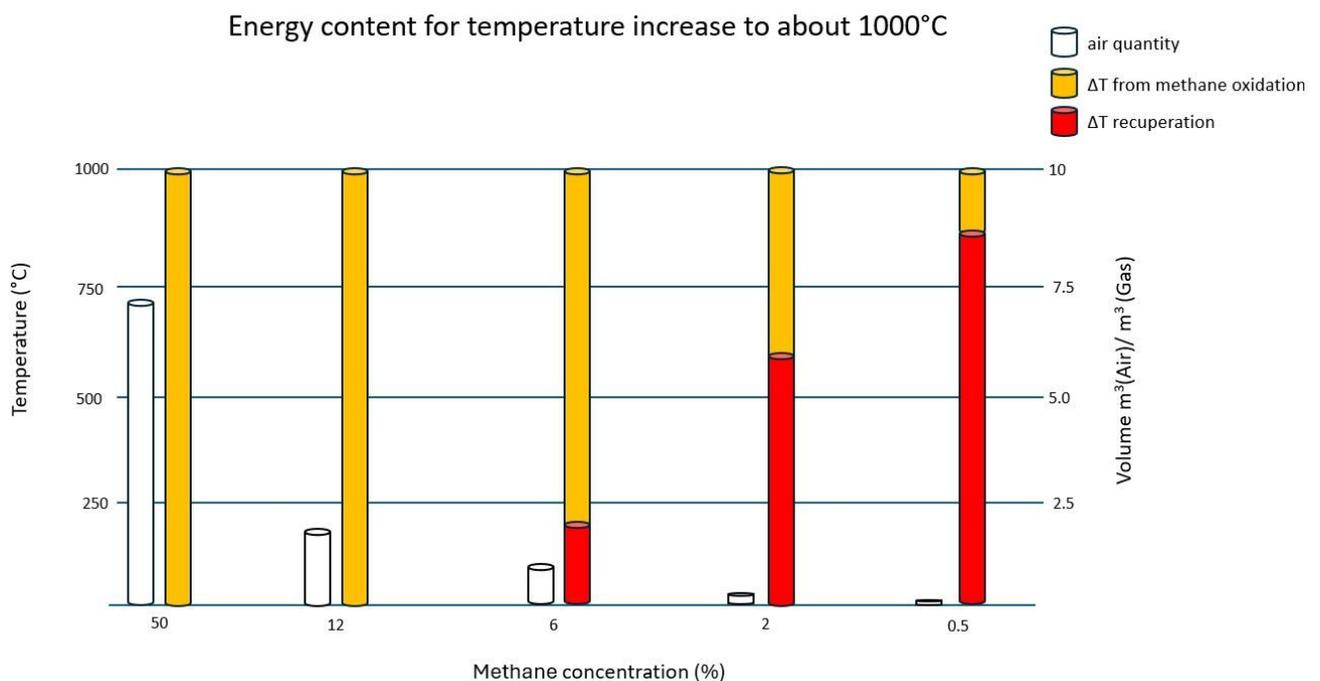


Figure 4: Required air volume ($\Lambda=1,5$) and partition of energy required to reach 1000°C (Temperature increase due to the oxidation of methane respective required recuperation from off gas)

In technical applications (e.g. flares), when methane concentrations are below approximately 12 vol. % in the fuel gas (oxygen free), stable and complete combustion at 1000°C can no longer be guaranteed without recuperation or the regeneration of heat. There are available technologies for concentration ranges below 12 vol. % that can lead to complete oxidation by recuperation and/or regeneration. Energetic conditions and the national regulations (e.g. residence times in the combustion process, temperature requirements, methane slip) need to be considered when selecting such a technology.

Table 9 presents the methane oxidation technologies that are suitable for specific methane concentration ranges in the fuel gas.

Table 9: Technologies suitable for certain methane concentration ranges (The limits differ for specific applications and individual specifications.)

Methane Oxidation Technology	Concentration of methane in fuel gas (oxygen free) (vol. %)	Remarks
Combined heat and power unit (CHP) CHP with lean gas fuel mixer	100-35% 20-50 %	Low methane concentrations make the startup procedure difficult, lean gas mixers can add more fuel gas than the conventional mixers for natural gas or biogas, so lower methane concentrations are possible.
Flare	25-100%	Oxygen (air) supply and cooling by air is achieved by natural ventilation
Flare with controlled air supply	12-100%	Oxygen (air) supply and cooling air is added by means of a controller; operation of the flare can be adjusted to changing conditions if needed (only relevant to applications with changing gas composition (e.g. landfills))
Oxidation system with heat recuperation to air supply	6-100 %	Heat recuperation and air supply can be adjusted to changing conditions if needed (only relevant to applications with changing gas composition (e.g. landfills))
Oxidation system with heat recuperation to air and fuel gas supply	(2) 3-100%	Heat recuperation and air supply can be adjusted to changing conditions if needed (only relevant to applications with changing gas composition (e.g., landfills))
Regenerative thermal oxidation	0.37 - 100	RTOs have a design concentration, so higher methane concentrations need to be diluted to that design point.

6.1 COMBINED HEAT AND POWER (CHP)

CHP units can be used for fuel gases with a methane content between 100 and 20 vol. %. Standard biogas or landfill CHP applications can be operated approximately until 35 vol. %; below this specific gas mixers need to handle the high-volume flows of the fuel gas. For mining gas and landfill gas such a mixer has been developed and a methane content of approximately 20 vol. % can be oxidised (Green Gas 2018).

Since the manufacturers of CHP units design the components for certain gas concentrations (e.g. natural gas or biogas characteristics), conventionally available CHP units are limited with regard to the usable gas composition due to their fuel and air supply requirements.

For methane contents that are significantly lower than the standard design for biogas or landfill gas, the fuel gas feed must be designed appropriately. Retrofit kits are available for this purpose, but not all manufacturers offer them, or they are not compatible with all systems. The start-up of the CHP might become difficult and also compliance with emission limits under these conditions must be checked.

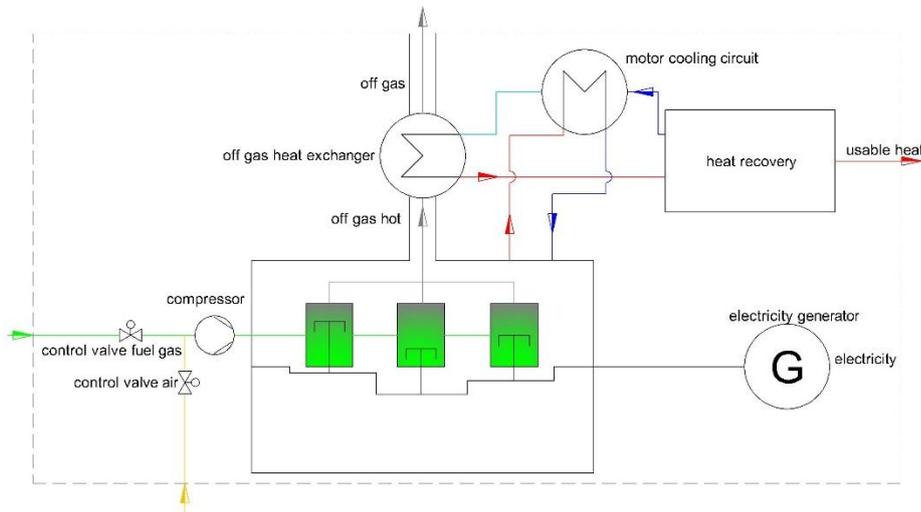


Figure 5: Technology principle combined heat and power plant

Another option is the addition of a lean gas into the air supply of a CHP. Here the concentrations need to be monitored closely, the manufactures of CHP need to be involved, and safety regulations need to be considered.

6.2 FLARE FOR HIGH CALORIFIC GASES

Standard flares for high calorific gases ($\text{CH}_4 > 25$ vol. %) have a simple design with an insulated combustion chamber, which is dimensioned according to the thermal requirements of the respective local regulations. The structure normally contains the following components:

- Gas burner for optimum mixing of fuel gas and combustion air
- Materials for burner and insulation: V4A, ceramic insulation, resistant up to $1,400^\circ\text{C}$
- Combustion air louvre
- Operation monitoring by means of UV sensor

The insulation is dimensioned in such a way that the combustion temperature of e.g. $1,000^\circ\text{C}$ can be safely reached and also maintained for the minimum retention time. The plant is temperature controlled; and the combustion air volume is adjusted via an air louvre. This control concept functions reliably down to approximately 25 vol. % methane in the fuel gas. Up to this operating point, at least 5 m^3 of combustion air is required per m^3 of fuel gas.

If the methane concentration falls below 25 vol. %, the control accuracy of the louvre dampers is no longer sufficient, so that either too little or too much air enters the combustion chamber due to the inaccurate setting of the combustion air volume. This then leads, alternately, to sub-stoichiometric operating conditions and to an excess of air, so that the combustion chamber setpoint temperature is undershot. Stable plant operation is no longer possible (Ramthun 2022).

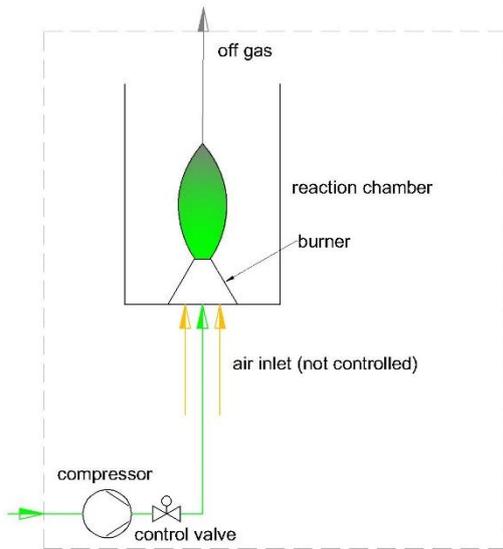


Figure 6: Flare concept

6.3 LEAN GAS FLARE - FLARE WITH CONTROLLED AIR SUPPLY

The modified flare for the treatment of gases with methane content from 100 to > 12 vol. % adopts the most important components of the standard flares, in terms of concept and design. The main components such as combustion chamber and burner, as well as the materials used are the same. However, the air supply needs to be controlled by an additional, infinitely variable combustion air blower to maintain the necessary temperature and oxygen supply in the combustion chamber stable. Operation monitoring is ensured by a combination of a UV sensor and a temperature sensor (Ramthun 2016). The precise regulation of the air supply prevents the system from entering a sub-stoichiometric range. The systems are applied in Germany at landfills, where landfill gas production has ceased, and the extracted landfill gas is increasingly diluted with nitrogen coming from air introduced into the landfill.

A company in Germany offers such a flare system with the flare having a specific surface burner based on a metal mesh. The gas to be treated is mixed with air and pressed through a hot metal mesh, on the surface of which the methane oxidation takes place.

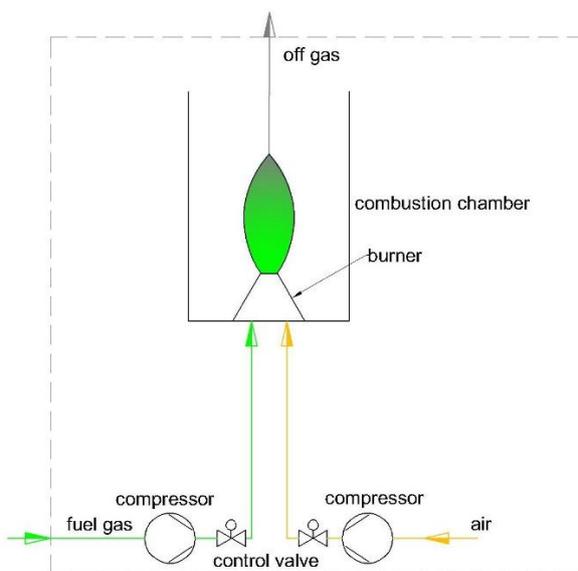


Figure 7: Lean gas flare

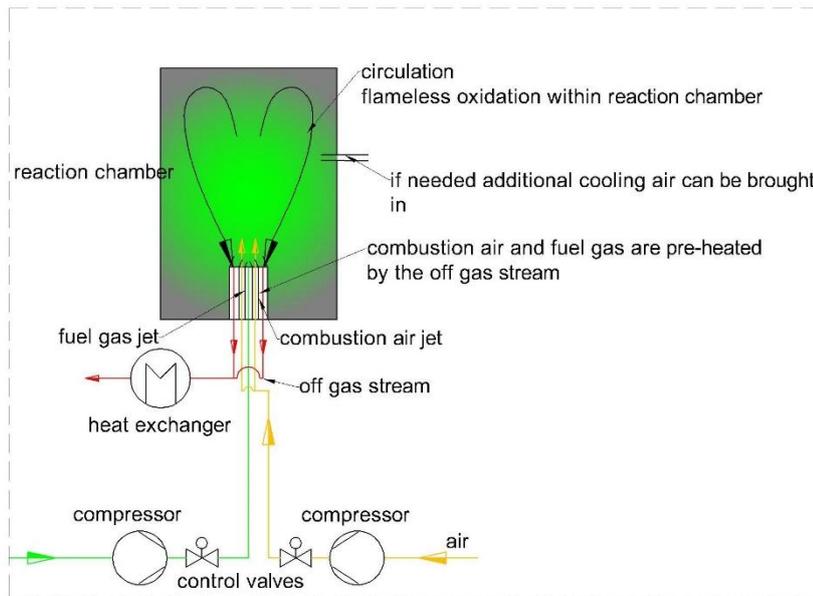


Figure 9: E-Flox burner

6.6 REGENERATIVE THERMAL OXIDATION (RTO)

In contrast to the treatment processes presented so far, regenerative thermal oxidation (RTO) is a discontinuous process. During treatment the gas flows through a ceramic heat exchanger bed. The incoming cold gas is heated while passing through the heat exchanger bed until the mixture oxidizes without a flame. The temperature increases and the heat in the downstream section is transferred to the heat exchanger bed. After a while the direction of flow is reversed to avoid the movement of the hottest combustion area out of the desired area in the heat exchanger bed. Depending on the cycle, heat from the oxidation is transferred to the heat exchanger material or the heat exchanger material transfers heat to the incoming cold fuel gas-air mixture. There are different designs; some have a combustion chamber between the heat exchangers, while in other applications the oxidation takes place directly in the ceramic bed.

Description of Cycles

The gas to be treated flows into the heat exchanger, heats up and oxidizes. In the process, the inflow area cools down, and output area heats up. Then the gas flow is stopped, and the flow is reversed. The cold gas now enters via the hot section of the previously output area.

When there are only two sections, one for input and one for output (2-chamber system), there is still unburned gas in the cold inlet area at the moment the cycles are switched. If this unburned gas is fed directly into the clean gas flow, this can lead to a short emission peak in the off gas. To avoid this, the section with the unburnt gas can be purged in a 3-chamber RTO with a partial flow of clean gas. The purge gas is recirculated into the fuel gas-air mixture and passes through the oxidation process. Since the heat losses in the RTO must be kept low, it is precisely dimensioned for a constant energy content in the air/fuel gas mixture. If the fuel gas does not contain oxygen or/and its energy content is too high energy content, air must be added to set the right conditions. The RTO has a narrow concentration range of methane (or energy content) in the gas mixture to the oxidation chamber. Consequently, if the energy content of the fuel gas (methane concentration) changes, the amount of air added must be adjusted. The flow rate can be varied within certain limits during operation of an RTO, with the lower limit being the necessary inflow velocity for uniform mixing in the ceramic bed, and the upper limit being the thermal capacity and the necessary residence time in the process. Since most industrial processes are oriented towards a stationary process, such adjustments of the air admixture are rather the exception (e.g. at old landfill sites with decreasing methane production).

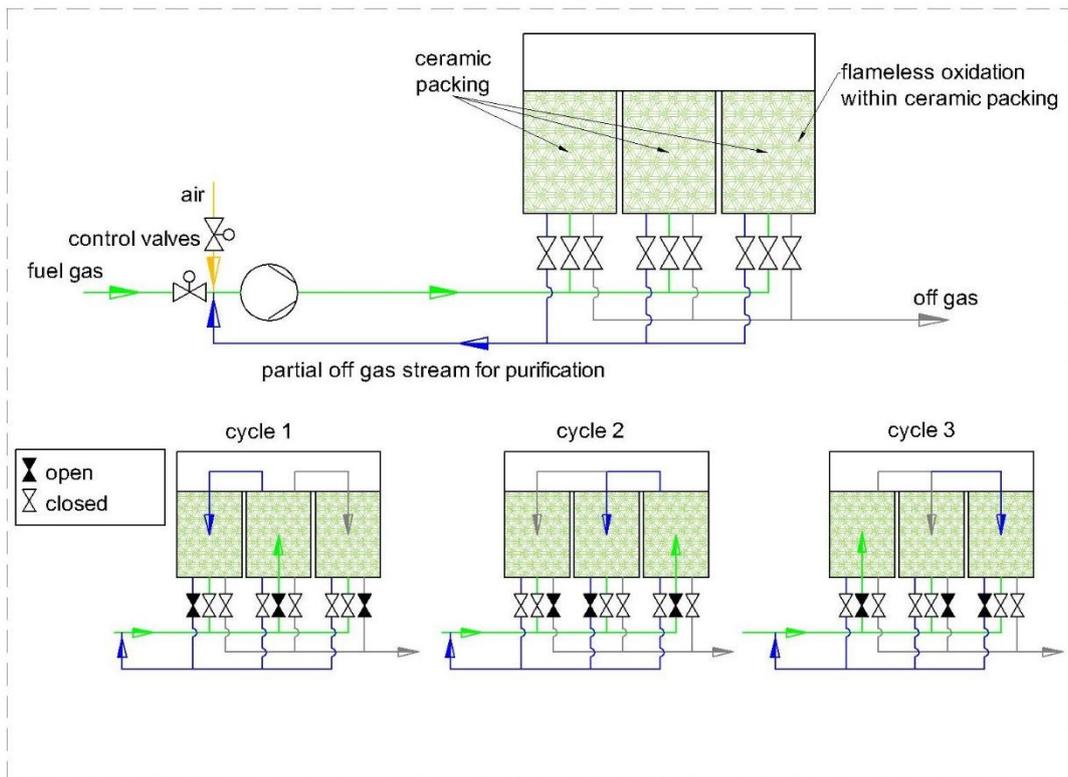


Figure 10: Regenerative Thermal Oxidation (RTO) Operating Cycles

For landfill gas oxidation applications, it is often necessary to design for a relatively wide range of methane flow rates and concentrations as the amount of methane formed decreases and the landfill gas becomes more diluted with air. Even if methane formation in the case of old landfills is declining, an RTO can be operated for a long period of time. For the treatment of off gases from biogas upgraders and CHP units, the RTO technology is the only technology capable of handling methane concentrations in the fuel gas below 2 vol. %, and it operates down to 0.37 vol. %.

Furthermore, the operation of an RTO in start-stop mode is energy-intensive, since the heat exchangers have to be preheated before start-up. This can be done by supplying high calorific fuel gas or by electrical heating. On the other hand, shutdowns of short durations are possible, as the regenerators cool down very slowly and can be brought back to temperature quickly.

Below a methane concentration of 0.37 vol. % in the raw gas and a capacity of 10 - 15 kW thermal power, an autothermic treatment (without addition of extra energy) can not be guaranteed. RTOs of this size are not standard, and it may be difficult to find manufacturers who are able and willing to produce such small units.

6.7 TECHNICAL APPLICATIONS FOR BIOLOGICAL METHANE OXIDATION

Methane oxidation biosystems (MOB) have been intensively investigated and are being progressively implemented in the field of waste disposal. The vast majority of applications are passive flow systems, or Passive Methane Oxidation Biosystems (PMOBs) (Nelson et al 2022). In this context, passive means that the treated gas is driven through the biosystem by natural processes, primarily changes in barometric air pressure (see Figure 11). As a result, process conditions such as flow rates and gas composition can hardly be controlled and can vary greatly, in particular when there are sudden changes in barometric pressure, which can rapidly increase or decrease the natural flow. In contrast, in active MOB, the gas is transported through the biosystem in a manner defined by design, with more control over process parameters (see Figure 12).

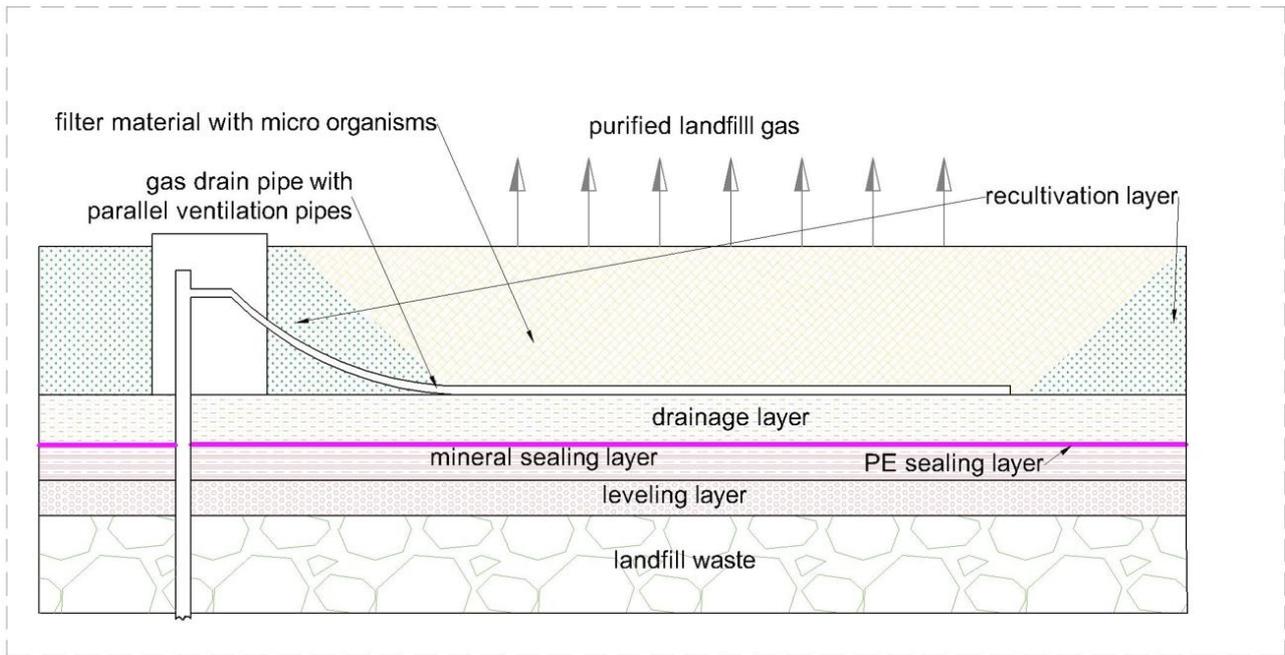


Figure 11: Methane oxidation window (passive system)

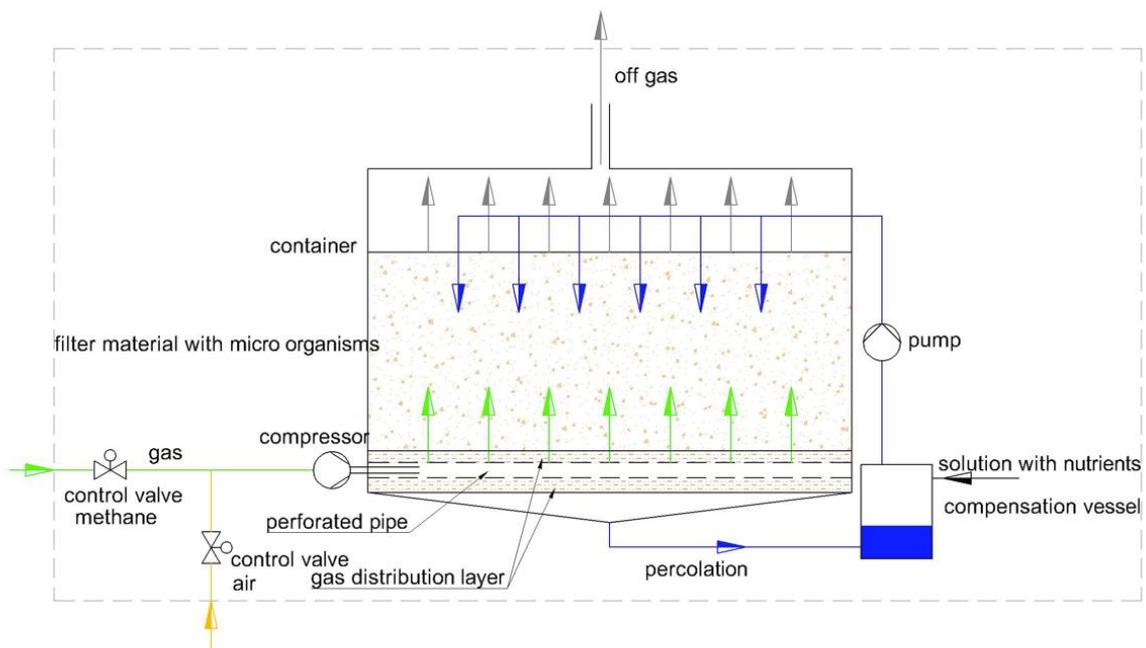


Figure 12: Methane oxidation filter (active system)

The performance of passive flow-through biosystems has been intensively investigated. Scheutz et al 2009 presents an overview of technologies and degradation rates of active and passive systems in a comprehensive review, whereas (Gebert et al. 2022) present design elements for the main types of biosystems, namely biofilters, bio-windows and bio-covers. Gebert and Pfeiffer (2017) propose a "standard oxidation capacity" for passive systems, which is 1 L CH₄ /m² filter area and hour (equivalent to 17 g CH₄/m²day or 6.2 kg CH₄/m²annum). This is adjusted for temperature, pore volume and water tension. Gebert et al (2022) provides further design parameter and recommendations for methane oxidation systems. Also, Gebert and Pfeiffer (2017) also mentions maximum oxidation rates for composts as filter material of up to 63 g/m²h; in Gebert et al (2022) 24-33 g/m²h is the maximum range reported.

Accordingly, the question arises as to how the oxidation performance of active systems can be optimised in comparison to passive systems to obtain higher degradation rates and thus smaller areas or filter volumes.

In contrast to passive systems, active systems can be integrated into technical processes and are significantly more efficient due to their controllability. Flow, moisture and, above all, the oxygen content in the filter can be controlled, so the filter volume is used more effectively. Most scientific publications focus on the maximum methane oxidation rates, whereas the long-term behaviour and the dependence on operating parameters is more difficult to assess. Haubrichs and Widmann (2006) report an increase in the degradation performance of the passive versus the active system by a factor of 5.5 ($5.1 \text{ g}/(\text{m}^3 \text{ filter volume h})$) to more than $28 \text{ g}/\text{m}^3 \text{ filter volume h}$). It must be noted here that the passive system used as a basis already has a very high degradation capacity, compared to the standard oxidation capacity mentioned above.

Streese and Stegmann (2003) carried out a long-term test with various filter materials and, based on the results, propose a dimensioning (flow rate) of approximately $1 \text{ m}^3/\text{m}^3 \text{ filter volume h}$ for optimum conditions and $0.4 \text{ m}^3/\text{m}^3 \text{ h}$ for the more conservative approach, i.e. "stable conditions". The "stable conditions" approach was calculated by adding a safety margin to the data determined from the test results. For the load per filter volume, about $17.3 \text{ g}/(\text{m}^3 \text{ h})$ (corresponding to $24 \text{ L CH}_4 /(\text{m}^3 \text{ h})$) is proposed for optimal conditions and $7.7 \text{ g}/\text{m}^3 \text{ h}$ ($10.6 \text{ L CH}_4 /\text{m}^3 \text{ h}$) for the "stable conditions" variant. The mean methane concentration in the gas was 2.5 vol.%. The degradation performances fluctuated very strongly over the test period.

Scheutz et al (2009) presents various studies on biofilters and states that the degradation efficiencies of the majority of the studies are in the range of $340 - 591 \text{ g CH}_4 (\text{m}^2 \text{ d})^{-1}$, which corresponds to $14.2 - 24.6 \text{ g CH}_4 (\text{m}^2 \text{ h})^{-1}$.

It should be noted that high degradation rates do not guarantee high degrees of degradation (or high degradation efficiencies). With increasing freight to the filter, it can be assumed that the degradation rate and capacities increase, but the degree of degradation decreases due to higher load. The impact is influenced by how the loading is ramped up and then how it is controlled. Since no clear design parameters can be derived from the data - in particular not with regard to achievable degrees of degradation - a loading in the order of magnitude of $18 \text{ g CH}_4 (\text{m}^3 \text{ h})^{-1}$ (corresponds to approximately $25 \text{ L CH}_4 (\text{m}^3 \text{ h})^{-1}$) is assumed as a potential design parameter in this report. On the basis of the available data, a considerable degree of degradation should be possible with this loading.

Another relevant parameter for filter dimensioning is the adjustable methane content in the raw gas to the filter. Here, a high value should be aimed for to achieve a high concentration gradient to the liquid phase and thus the highest possible absorption of the poorly soluble methane into the liquid phase. Another option to optimize the absorption is the design of the system, in particular the residence time of the gas in the system. Explosion protection plays also role for limitations here. If classification of explosion zones 1 or zone 0 are to be avoided in the filter, then an explosive mixture in the raw gas to the filter must be securely avoided. Depending on the technical design and legal requirements, a maximum value in % LEL (lower explosion protection limit; e.g. 40 % LEL, i.e. max. 1.8 vol.% CH_4) must be observed.

Furthermore, the flow rate through the system and the resulting residence time must be considered, since this primarily limits the transfer of the methane into the liquid phase. The real residence time is determined by the pore volume, which, however, changes continuously. Furthermore, it must be taken into account that when organic-rich materials are used for methane oxidation layer, its degradation with time reduces the active volume of the methane oxidation layer and thus the retention time. Here it must be ensured that the design value is maintained on average over the operating period. According to Streese and Stegmann (2003), $1 \text{ m}^3/(\text{m}^3 \text{ h})^{-1}$ is assumed as a guide value for the filter, i.e. pure filter volume without consideration of the pore volume.

For the following operation, due to safety issues a maximum methane concentration of 1.8 vol.% (40 % LEL) is assumed for the operation of a methane oxidation filter. This results in the design values shown in Table 10 for a load of 1 m³CH₄/h and the assumptions made:

Table 10: Dimensions for active methane oxidation biosystems

Load methane	Mixed gas to the filter at 1.8 % CH₄(m³)⁻¹	Size of filter according to specification Total flow rate 1 m³(m³h)⁻¹	Size Filter according to specification Load Filter 18 g CH₄ (m³×h)⁻¹
1 m ³ CH ₄ h ⁻¹	55.6 m ³ h ⁻¹	55.6 m ³	40 m ³

The dimensioning of the methane oxidation filter is based on a design value. With higher loading, as described above, it can be assumed that the absolute degradation performance increases, but the degree of degradation decreases. In general, a methane oxidation filter can be operated, within the limits of safe operation, with a high range of flow rates and concentrations for a short time. Especially during (re)start-ups or protective degassing, this can be helpful. In the long term, however, the operating data should correspond to the dimensioning variables.

If the filter is operated at lower flow rates and loading, a higher retention time results, which should have a positive effect on the oxidation performance. However, a uniform flow through the filter must be ensured no matter what the flow rate. This can only be achieved by incorporating unsaturated flow of moisture into the design phase (Ahooghalandari et al. 2018, Gebert et al. 2022). That is, moisture affects gas migration, insofar as it can cause pore blockage at the interface between the gas distribution layer and methane oxidation layer. Pore blockage decreases the actual volume where methane oxidation can occur.

6.8 METHANE EMISSION REDUCTION THROUGH UTILIZATION OF CO₂

Within biogas upgrading, if the separated carbon dioxide is to be further utilized as a product, the carbon dioxide-rich off-gas stream must also be further upgraded to meet the quality requirements for marketable biogenic CO₂ in the food industry, for example. As a result of this process, an off-gas stream is produced that contains the concentrated methane slip and thus has higher methane concentrations than the off-gas of the biogas upgrading process. This off-gas stream can then be recycled back into the inlet of the first biogas upgrading process. This in turn requires a larger upgrading capacity, and the closed system reduces the methane emissions next to zero.

The technical requirements for the upgrading of CO₂ are similar to biogas upgrading. Therefore, the upstream processes should not add gases (i.e. upgrading which uses strip air) since this adds volume and components to be treated.

The CO₂ -containing off gas stream from the upgrading plant is temporarily stored, if necessary, and conveyed to the CO₂ liquefaction plant. The steps are shown in Figure 13. In a first step, water-soluble components can be removed from the gas by means of a scrubber. The pressure of the gas to be treated is increased by means of a compressor, and impurities and residual moisture are removed by means of subsequent activated carbon filter and dryer units.

This cleaning step is followed by a CO₂ condenser, where the actual liquefaction takes place. The coolant liquefaction system provides the necessary condensation capacity. A compressor increases pressure and temperature in the gaseous refrigerant. The subsequent refrigerant condenser liquefies the high-temperature gas. The condensation heat of the gaseous refrigerant is transferred to a coolant through a heat exchanger. The refrigerant now evaporates in the CO₂ condenser and thus provides the required cooling capacity to liquefy the incoming CO₂ gas.

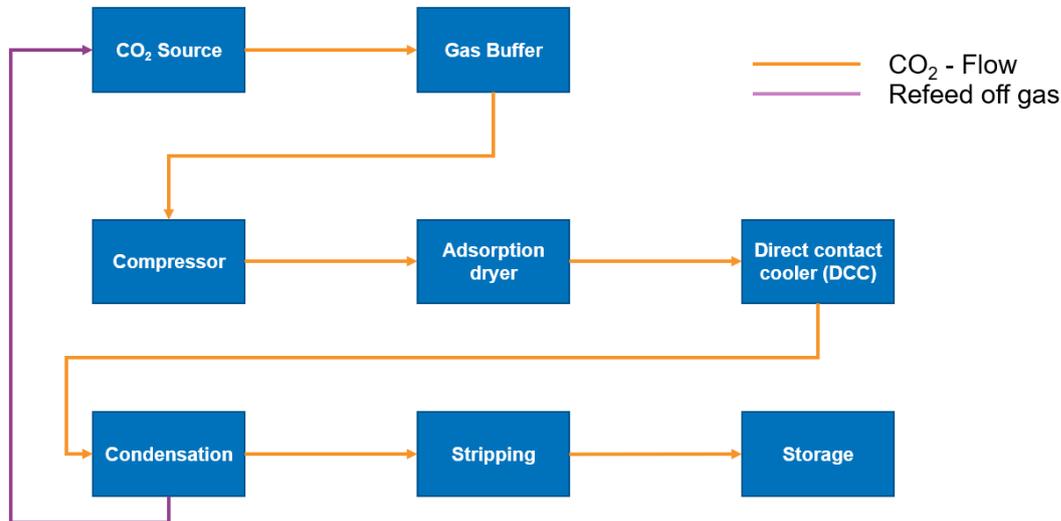


Figure 13: Process flow of CO₂ purification and liquefaction (Lehmann et al. 2024)

The liquefaction of the CO₂ gas takes place at approximately -24°C. The stripper or a subsequent reboiler removes the last non-condensable impurities (N₂, O₂ and CH₄) from the liquid CO₂. The resulting off-gas stream with non-condensable components can be recycled to the first biogas upgrading system. The stripping technique allows the methane contained in the CO₂ to be returned to the upgrading process and almost 100% pure liquid CO₂ to be produced. The liquid CO₂ is conveyed to an insulated storage tank and stored there until it is removed.

The utilization of the CO₂ allows not only the physical utilization. In case of long-term storage of biogenic CO₂ the system can even provide negative emissions as CO₂ originating from organic material comes from the atmosphere and storing it means removing it from the atmosphere.

7 Energy balances and possibilities for utilization of excess energy

The oxidation of methane is an exothermic reaction. In CHP units, the available energy can be converted into electrical energy and heat. At lower methane concentrations, the energy contained in the lean gas (e.g. below 20 vol. % CH₄) is not sufficient to provide electrical energy economically with available technology and current economic conditions. For example, available technologies such as Stirling engines can convert the gas, but are not often used due to economic reasons.

If heat is to be utilized, the quantity of heat and temperature level are the decisive factors for economic utilisation. While the quantity of heat is determined by the methane volume flow into the system and the losses from the conversion process, the temperature level depends on the gas composition, particularly the methane concentration, and the losses in the process.

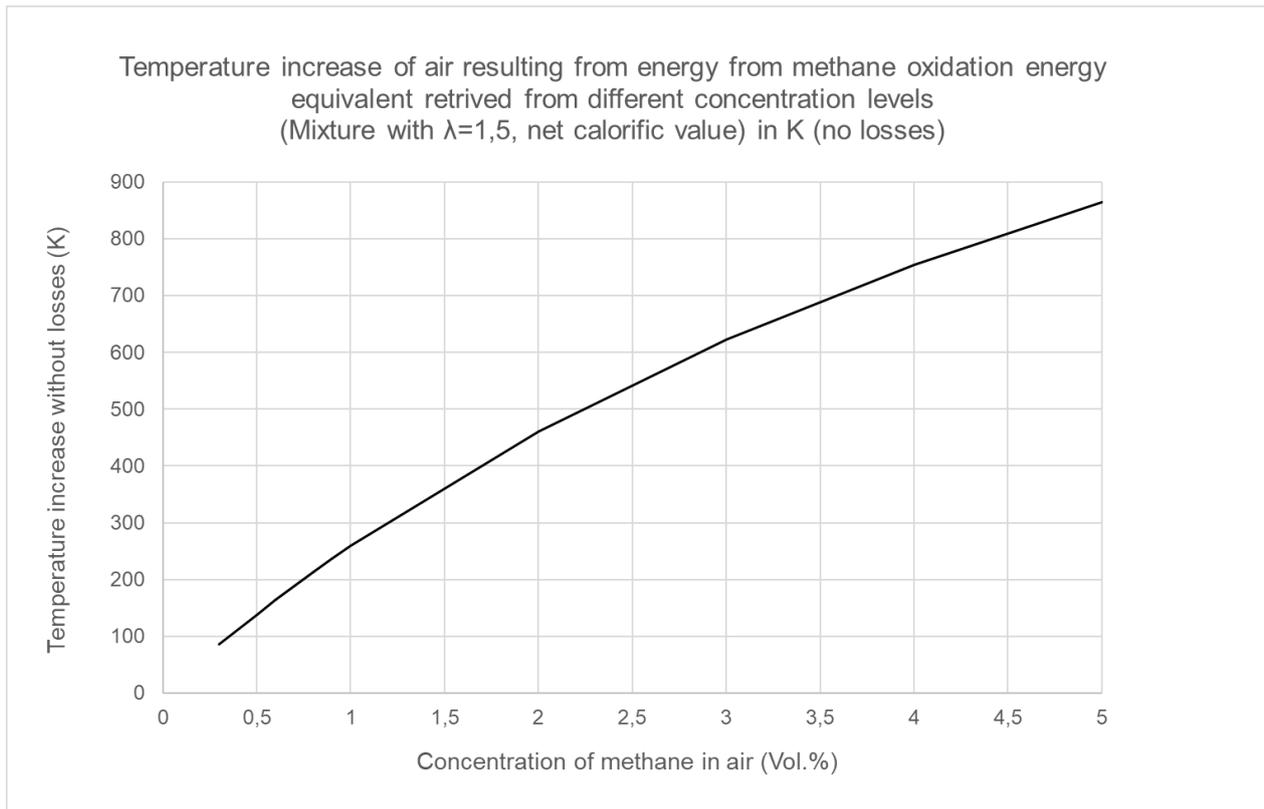


Figure 14: Temperature difference of air resulting from energy equivalent of different methane concentrations

It can be seen from Figure 14 that the required temperature increase for a complete oxidation (e.g. up to 1000°C) cannot be achieved at low methane concentrations. At energy equivalents resembling concentrations below 1 vol. % methane, the temperature increase resulting from oxidation is very low.

The oxidation process itself runs at higher temperatures due to the heat recuperation or regeneration, but the usable temperature is defined by the temperature level of the off gas. This temperature level is defined by the energy content of the gas mixture going into the oxidation process and the gas composition. Assuming that energy from methane oxidation heats up air, the available temperature increase is shown in Figure 14. As these are ideal conditions, without consideration of losses during the process, the practically available temperature is accordingly lower.

The lower the temperature of the off gas, the fewer application options are available for heat utilisation. Since even low temperature applications such as residential heating systems require a certain temperature level ($T > 30 - 55^{\circ}\text{C}$), the usable heat quantities become smaller as the flue gas temperature decreases. The lower the methane concentration in the fuel resp. raw gas, the lower the usable heat quantity. Since future energy prices are difficult to predict, it should at least be mentioned that the low temperature heat could also be upgraded by means of heat pumps. Air-to-Air heat pumps used for heating homes could be efficient with a constant heat supply such as the off gas of an RTO represents, in particular in the cold season of temperate zones. The application of heat pumps would make larger quantities of available low-temperature heat usable.

8 Economic analysis - case: RTO applied to off gas from upgrader

An economic analysis of the costs of methane emission reduction have been calculated for the application of Regenerative Thermal Oxidation (RTO) technology to the off gas of a biogas upgrader. This off-gas would typically have a concentration of at least 0.5 vol. % CH₄, which means that no additional heat supply for normal operation of an RTO is needed. Figure 15 shows the size distribution of upgrading plants in Europe and apparently many plants are operational in the 50 - 250 m³/h size range, few below. 50-250 is quite a large span for the volumetric capacity, and, it is assumed that more upgrading plants will be at the upper end of this range.

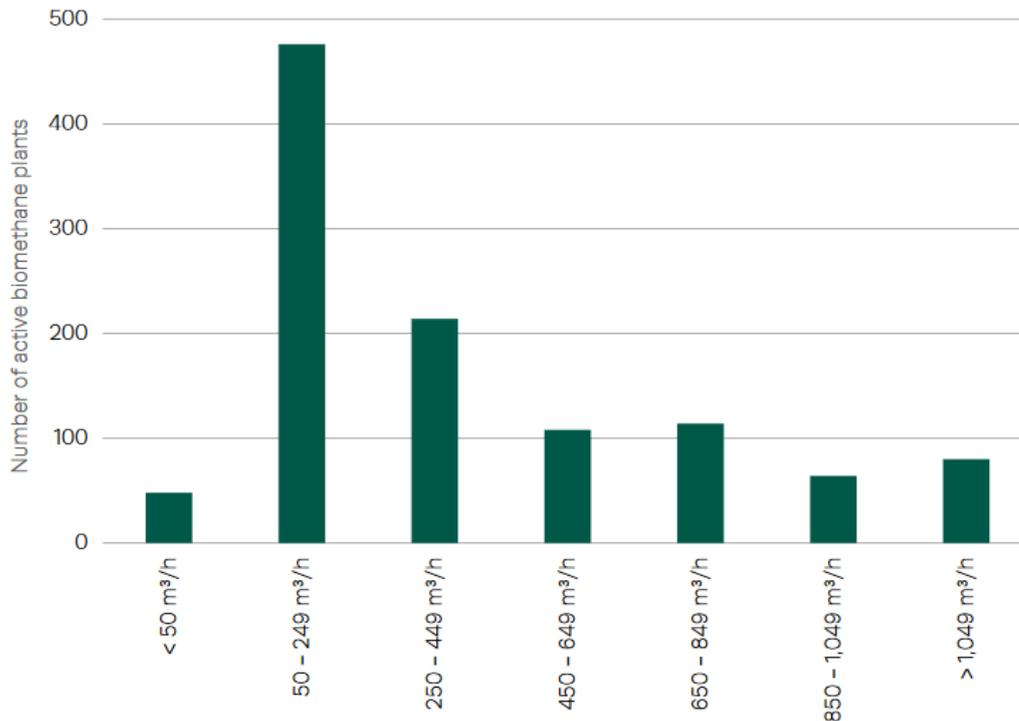


Figure 15: Plant size distribution in Europe (Feed in capacity, m³CH₄/h). (EBA Statistical report 2022)

This economic analysis is based on a rather small-scale facility - the lower range of plants in Europe, which treat between 45 and 122 m³ of off gas flow on an hourly basis. The investment costs for the RTO were assumed to be 330,000 €. Site specific costs such as commissioning, integration into the overall plant control system, operating permits and approvals are not included as they vary significantly when comparing an RTO retrofit with a new plant construction including the RTO.

It is assumed that the biogas composition is 55 vol. % CH₄ and 45 vol. % CO₂. This is decisive of the calculation of the amount of off-gas, which can be treated in the given RTO. The RTO has a range of operation to cover both capacity scenarios (see Table 11), however usually the RTO would be designed for a certain throughput. The costs for specifically designed plants should not deviate to a large extent, in this small size range. The available information is for a compact version of an RTO treatment facility, and according to economy of scale the costs for small scale plants represent the upper end of specific costs. RTOs can easily be scaled-up for larger biogas plants. The facility consumes 28,000 kWh of electricity on a yearly basis. A price of 0.08 €/kWh for electricity is assumed. Maintenance is provided through quarterly service checks at a cost of 5,800 € per annum. There is no information available to indicate that the operating costs change within the range of the plant's volume limits. Consequently, costs are the same in both calculations. However, the upper limit of the volume allows for a higher methane production than at the lower limit, which reduces the total costs per m³ CH₄ produced.

Table 11: Exemplar of cost calculation for 2 different RTO plant throughputs

Biogas flow	m ³ /h	100	222
Biomethane flow	m ³ /h	55	122
Off gas flow (CO ₂)	m ³ /h	45	100
Methane slip	%	1	1
Methane slip	m ³ /h	0.55	1.2
Methane emission reduction	m ³ /y	4,818	10,707
Total investment	€	330,000	330,000
Average lifetime	years	15	15
Interest rate	% p.a.	3	3
Annual capital costs	€/y	27,643	27,643
Electricity	€/y	2,260	2,260
Maintenance	€/y	5,800	5,800
Total costs	€/y	35,703	35,703
Abatement costs	€/kWh CH ₄ total production	0.007	0.003
Abatement costs	€/ton CO _{2eq} (GWP 28)	378	170

The calculation is based on the assumption that one unit is operated with a higher throughput and the resulting costs vary accordingly. This highlights the economy of scale, since in this plant size range the capital and operating costs for the RTO do not vary much in proportion to the throughput - but the abatement costs per unit of throughput do.

Under these given conditions the minimum abatement cost for the methane oxidation in the RTO is minimum 170 €/t CO₂e. While this cost seems high, it is reasonable and competitive when compared with the calculated abatement costs per t CO₂ that were, until Q3 2023, above 200 € for the transportation sector in Germany (Klima Quote 2024; it should be mentioned that this price dropped in 2024 due to sudden massive imports of low-priced biofuels from Asia). For very small biogas upgrading plants, methane emission reduction using RTO technology is expensive, but the abatement costs drop quickly as the plant capacity increases due to economies of scale. The case presented here represents the minimum capacity of the technology available. In general, the operation costs are higher at lower scale as some costs are independent of size (e.g. maintenance personal costs).

Regarding the impact on the biomethane production costs the calculation presented indicates that the addition of an RTO has limited impact, i.e. 0.3-0.7 ct€/kWh, compared to selling prices of e.g. 6-9 ct€/kWh in Germany.

The above analysis represents the lower limit of a potential RTO operation. Due to economies of scale, the unit costs for methane emission reduction at larger plants would be lower.

9 Selected example plants

9.1 CO-COMBUSTION OF LEAN GASES IN CHP UNITS

Projekt: Zeche Erin, Germany

Green Gas Germany has been operating a CHP plant with a capacity of 1.4-MW in Castrop-Rauxel, with a CH₄ content below 20 vol.%. The lean gas comes from a closed mining area (Zeche Erin). The plant was commissioned in 2006 and has installed the patented lean gas kit.



Figure 16: Green gas CHP installation for mining gas (Source: Green Gas 2024)

9.2 LEAN GAS COMBUSTION SYSTEM - CHC

A lean gas combustion system is located at a landfill in Southwestern Germany (near Reutlingen). CHC is the product name of the company Lambda (in Germany) and is a commonly used system for lean gas treatment at old landfill sites in Germany. The collected gas from the landfill is incinerated in the system, which has a capacity range from 25-250 kW (approximately 2.5-25 m³CH₄/h). Due to this wide operating range, the operation of the system can be adjusted to the decreasing gas production of the landfill. The incineration temperature is 1000-1200 °C, the minimum methane content in the fuel gas is 12 vol.% and the maximum 100 vol. %.



Figure 17: Lean gas combustion system (“CHC”) (Source: Lambda 2024)

9.3 MEMBRANE SEPARATION WITH RTO AT WESTHEIM-BIETIGHEIM

Approximately 760 m³ of raw biogas with an average methane content of 55 vol.% is upgraded into biomethane at the site using membrane separation. The methane content after upgrading is 96 vol.%. The plant's feed-in capacity is 550 m³ of biomethane per hour, but on average, it operates at a rate of 435 m³ of biomethane per hour. The methane slip, based on the amount of methane fed into the upgrader, averages 1.1 vol.%. The CO₂-rich, low-methane flue gas stream is treated in a 2-chamber RTO from the German company IGS. The temperatures in the combustion chamber are between 850 and 950 °C. The clean gas temperature at the stack outlet is approximately 70 °C. The cycle time for switching the chambers is 100 seconds. The cleaning performance of the RTO is 99.6 vol.%, i.e. methane reduction in the off-gas. The plant was built by the company Venjakob (RTO) in collaboration with ETW (upgrading unit).

Table 12: Specifications of RTO at Westheim Bietigheim

	Biogas	Biomethane	Feed gas to RTO	Clean gas after RTO
CH ₄ (vol.-%)	58.10	95.70	1.10	0.004
CO ₂ (vol.-%)	41.90	1.52	n.a.	25.10
O ₂ (vol.-%)	0.00	0.00	n.a.	15.20
Throughput (m ³ /h)	636	385	271	814



Figure 18: Upgrading plant (Membrane) with 2-chamber-RTO (source Biogutenergie 2024; DBFZ)

9.4 LEAN GAS FLARE WITH RECUPERATIVE PREHEATING OF FUEL GAS AND AIR

The plant, manufactured by Göbel Energie- und Umwelttechnik Anlagenbau GmbH, is located in Bavaria, Germany on a closed landfill site. The plant processes 10-100 m³/h of landfill gas with a methane content of 60 - 4 Vol.%. The maximum capacity to the plant 100 kW_{th}. The incoming air and the fuel gas can be heated up to 750 °C if necessary. In standard operation the off gas is used to recuperate incoming air and fuel gas, during start up the system is warmed up by adding propane. The operational capacity range of the combustion system is 1:10, the combustion temperature is min 1000 °C and the retention time of the gas in the combustion chamber ≥0.3 sec.



Figure 19: Plant at Posthof (© Göbel)

9.5 METHANE OXIDATION FILTER AT A LANDFILL SITE (GERMANY)

The methane oxidation filter at a landfill site in Germany is used to treat the collected landfill gases. As the landfill has been closed for a long time, it only yields very small amounts of methane (approximately $2.0 \text{ m}^3\text{CH}_4 \text{ h}^{-1}$) with a low methane concentration (3.1 %), and a methane oxidation filter is the most economical solution. The methane oxidation filter has been in operation since the end of 2021, designed in container construction (reinforced HDPE) with 2 closed containers connected in series. The area of a container with the dimensions of $9.88 \times 2.26 \text{ m}$ is approximately 22 m^2 . The filter material is placed on a grate floor, the filter itself consists of an approximately 50 cm thick layer of root wood and an overlying layer of approximately 1.4 m mixture of bark mulch/spruce chips (50/50). The filter volume of a container is thus approximately 42 m^3 . The filter is equipped with a sprinkler system and a leachate collection system. With a methane volume flow of $2 \text{ m}^3\text{CH}_4 \text{ h}^{-1}$ (raw gas to the filter: $112 \text{ m}^3 \text{ h}^{-1}$) the loading rates shown in Table 13 result:

Table 13: Specifications methane oxidation filter

Filter volume:	84 m^3	Flow rate	$112 \text{ m}^3 \text{ h}^{-1}$
Filter area	44 m^2	Load:	$2 \text{ m}^3\text{CH}_4 \text{ h}^{-1}$
Assumed pore volume	50%		
Flow rate per unit area	Approx. $2.5 \text{ m}^3(\text{m}^2\text{h})^{-1}$	Retention time total volume	0.77 h
Flow rate by volume	Approx. $1.3 \text{ m}^3(\text{m}^3\text{h})^{-1}$	Residence time only pore volume	0.38 h (23 min)
Area load	Approx. $45 \text{ L CH}_4(\text{m}^2\text{h})^{-1}$	Approx. $31.5 \text{ g CH}_4(\text{m}^2\text{h})^{-1}$	
Volume load	Approx. $24 \text{ L CH}_4 (\text{m}^3\text{h})^{-1}$	Approx. $17 \text{ g CH}_4(\text{m}^3\text{h})^{-1}$	



Figure 20: Methane oxidation filter

The degradation performance of the filter shows a highly seasonal behaviour, peaking in the summer with 50°C temperatures in the filter material. In winter the methane conversion was very low at temperatures below 10°C in the air and filter. The gas might not be distributed evenly in the filter, so not all parts of the filter might be active. In addition to the oxidation taking place in the filter, the high extraction rate prevents emissions to be released from the landfill and causes methane oxidation to take place in the landfill due to air suction into the landfill.

9.6 METHANE OXIDATION BIOSYSTEM AT THE KITCHENER LANDFILL | MCLENNAN PARK

This ongoing pilot project is taking place at the old Kitchener Landfill (1958-1978) in Ontario (Canada), which has since become the McLennan Park (Nelson et al 2022). Landfill gas is still being pumped from the site. Two versions of this Methane Oxidation Biosystem (MOB) were installed to test the capacity of two types of filter material to minimize the impact of fugitive methane emissions from the site. Three LFG wells were installed at a depth of 4 m and deliver up to 50 m³/day of LFG (60 vol.% CH₄, 30 vol.% CO₂) to the biosystem. Some peaks of up to 128 m³/day of LFG were recorded during periods of low atmospheric pressure.

The MOB is contained in a 9.5m x 3.0 m x 1.2m plywood box on an 8% slope, supported with 2 inch x 4 inch framing and outer bracing. Water collected downslope is drained out through a heat-traced 4 inch p-trap. The design included a ¾-inch gravel base that serves as gas distribution layer (GDL). The GDL was placed upon a geotextile layer, which, in turn, was placed above the geomembrane. The pipe distribution system integrated into the GDL distributes the gas loading evenly throughout the surface of the MOB. Two transitional layers were placed above the GDL: ¼-inch gravel and sand. A heating trace system was installed in the sand layer to maintain optimal temperatures in the methane oxidation layer (MOL) throughout the colder months in Southwestern Ontario. The LFG influx was controlled by a mass flow meter.



Figure 21: Aerial view of the MOB (version 1) (Nelson et al 2022)

First version of the MOB: MOL composed of a mix of Compost and plastic pellets (SEPT. 2020 - Dec. 2021)

The first version of the MOB was constructed in October of 2020, which required the installation of the wooden frame, lining, drainage system, LFG wells, and monitoring equipment. A mixture of 50% domestic compost and 50% plastic pellets (volume basis) was used to fill the 90 cm thick (89 cm after settlement) MOL. Gas sampling probes were installed, water content probes and tensiometers (suction) were installed at several depths. The gas probes allowed to build gas profiles at three areas of the MOB, namely upstream, midstream and downstream; whereas the water content probes allowed verification of pore occlusion and the tensiometers allowed verification of the exactness of the initial design. The flowrates (methane loading) varied from season to season, between 0.25 to 5.33 m³/h. Surface flux measurement were conducted frequently (at least once a week) and showed an oxidation efficiency between 73% to 100%.

Table 14: Specifications of methane oxidation filter Kitchener (first version)

Filter volume:	25.65 m ³	Flow rate	0.25 – 5.33 m ³ h ⁻¹
Filter area	28.5 m ²	Load:	0.15 – 3.20 m ³ CH ₄ h ⁻¹
Assumed pore volume	60%		
Flow rate per unit area	0.009 – 0.187 m ³ (m ² h) ⁻¹	Flow rate by volume	0.010 – 0.208 m ³ (m ³ h) ⁻¹
Retention time total volume	102.6 – 4.8 h	Residence time only pore volume	61.6 – 2.9 h
Area load	5.3 – 112.3 L CH ₄ (m ² h) ⁻¹	3.8 – 80.2 g CH ₄ (m ² h) ⁻¹	
Volume load	5.8 – 124.8 L CH ₄ (m ³ h) ⁻¹	4.2 – 89.1 g CH ₄ (m ³ h) ⁻¹	

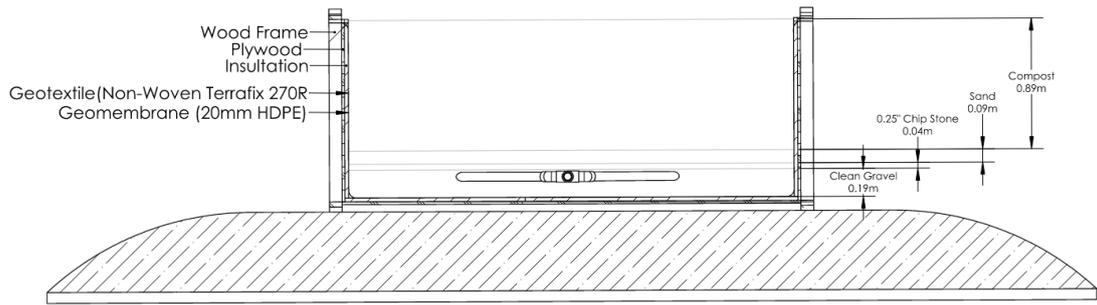


Figure 22: Section of the MOB (version 2) (Nelson et al 2022)

Second version of the MOB: MOL composed of a mix of Sand and Compost (SEPT. 2022 - Dec. 2023)

A second iteration of the MOB was implemented in May of 2022 using the same original frame, lining, drainage system, LFG wells, and monitoring equipment. A second line of heating trace was added 15 cm above the transitional sand layer, given that the optimal temperature regime for methane oxidation could not be maintained during the first iteration. In this iteration a 66% sand and 34% compost (volume basis) was used to fill a 50 cm thick MOL. Gas sampling and soil monitoring probes (water content and suction) were installed at several depths. They served the same purposes indicated above (first version of the MOB). Currently, flowrates have been varying between 0.21 to 1.88 m³/h. The corresponding oxidative efficiencies range between 97 and 74 vol.%.

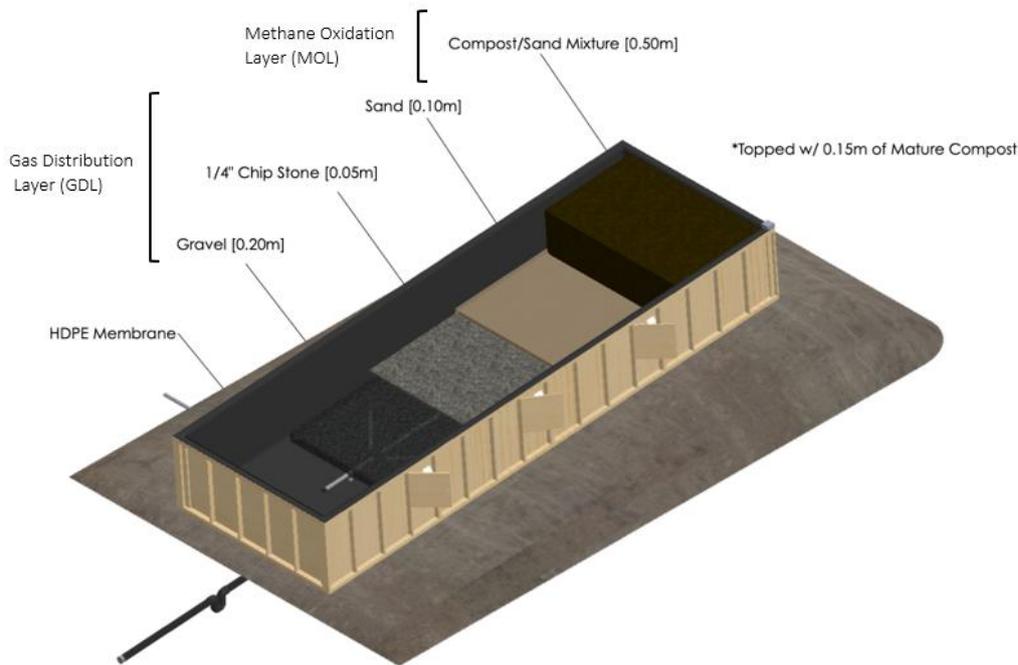


Figure 23: Top-Level Model of the Completed MOB (version 2) (Nelson et al 2022)

Table 15: Specification of methane oxidation filter Kitchener (second version)

Biosystem volume:	14.25 m ³	Flow rate	0.21 – 1.88 m ³ h ⁻¹
Biosystem area	28.5 m ²	Load:	0.13 – 1.13 m ³ CH ₄ h ⁻¹
Assumed pore volume	48%		
Flow rate per unit area	0.007 – 0.066 m ³ (m ² h) ⁻¹	Flow rate by volume	0.015 – 0.132 m ³ (m ³ h) ⁻¹
Retention time total volume	67.9 – 7.6 h	Residence time only pore volume	32.6 – 3.6 h
Area load	4.4 – 39.6 L CH ₄ (m ² h) ⁻¹	3.2 – 28.3 g CH ₄ (m ² h) ⁻¹	
Volume load	8.8 – 79.2 L CH ₄ (m ³ h) ⁻¹	6.3 – 56.5 g CH ₄ (m ³ h) ⁻¹	

9.7 METHANE OXIDATION BIOSYSTEM AT THE COMPLEXE ENVIRONNEMENTAL DE SAINT-MICHEL (MONTREAL LANDFILL | FRÉDÉRIC-BACK PARK)

The methane oxidation biosystem at the Complexe Environnemental de Saint-Michel (CESM) is a pilot project used to treat diluted landfill gas that would otherwise be vented directly to the atmosphere. The CESM is an old landfill that is currently being converted into a park (Frédéric-Back Park) in the heart of a neighbourhood in Montreal. While the site is already equipped with flares and a gas capture and utilization system, in the 2000s, a survey revealed that a portion of the landfill gas was migrating laterally near the boundaries of the site. To prevent landfill gas from migrating into the neighbouring residences, wells were installed in a trench near one of the boundaries to the landfill. However, the landfill gas in this area did not contain enough methane to be flared or for energy conversion. Hence this biosystem project was created to mitigate emissions from this trench. This 50 m² pilot project biosystem treats about 50 cfm (85 m³ h⁻¹) of depleted landfill gas containing between 0.5 to 4 vol.% methane.

The biosystem has been in operation since September 2021 (Almeida et al. 2024). Its walls were built of concrete blocks covered with insulating foam, a separation geotextile, and a geomembrane. The biosystem is divided into the two following layers: the gas distribution layer (GDL) and the methane oxidation layer (MOL). The 50 cm thick GDL was constructed using coarse gravel and progressively finer gravel to sand to act as filter. HDPE pipes were placed within the gravel layer to load depleted landfill gas into the MOB. A heating trace was installed on top of the sand layer. The approximately 1 metre thick MOL is composed of a 50/50 mixture by volume of compost and woodchips. The total volume of the biosystem is close to 76 m³.

Table 16: Specifications of methane oxidation system in Montreal

Biosystem volume:	76 m ³	Flow rate	85 m ³ h ⁻¹
Biosystem area	52 m ²	Load:	0.42-4.25 m ³ CH ₄ h ⁻¹
Assumed pore volume	66%		
Flow rate per unit area	Approx. 1.62 m ³ (m ² h) ⁻¹	Flow rate by volume	Approx. 1.12 m ³ (m ³ h) ⁻¹
Retention time total volume	0.89 h (app. 54 min)	Residence time only pore volume	0.59 h (app. 35 min)
Area load	Approx. 8.1-81 L CH ₄ (m ² h) ⁻¹	Approx. 5.8-58 g CH ₄ (m ² h) ⁻¹	
Volume load	Approx. 5.6-55.9 L CH ₄ (m ³ h) ⁻¹	Approx. 4-40 g CH ₄ (m ³ h) ⁻¹	
Efficiency	80-98%		



Figure 24: Methane oxidation filter in Montreal, QC (From A. Cabral)

9.8 LIQUEFACTION OF CO₂

Recycling Energie AG in Nesselbach, Switzerland processes 300 tons of food waste from restaurants, hospitals and residential facilities and turns this into biogas. Approximately 800 m³/h of the raw biogas is then upgraded and fed as natural gas substitute into the gas grid. The annually injected biomethane equals an energy equivalent of 35 GWh/a.

Biogas upgrading produces an off-gas, which contains mainly biogenic CO₂ and a residual proportion of methane, the methane slip. In general, such waste gas streams from upgraders are either combusted to reduce methane emissions or they are discharged into the environment, depending on the local regulations. Since March 2023 in Nesselbach the off-gas is treated further to turn the CO₂ into a commercial product. The CO₂ cleaning and liquefaction plant is one of only a few plants in Europe to provide biogenic CO₂ of a food grade quality. CO₂ is an important base product for the chemical and food industries. Large quantities of biogenic and therefore renewable and emission neutral CO₂ are needed to substitute fossil CO₂ which is currently the standard within the industries. Beside such carbon capture and utilization (CCU) options, there might be an increasing market for carbon capture and storage (CCS) technologies, where the CO₂ is stored underground.

The CO₂ upgrading and liquefaction plant is operated by a separate company, CO₂ Energie AG. The liquefaction plant produces up to 4,000 tons of food-grade liquid CO₂ per year. Industrial gases company, Messer Schweiz AG, collects the CO₂ three times a week with a tanker and sells it to manufacturers such as producers of dry ice or carbonated drinks. CO₂ Energie AG generates further income for the operation of the plant through the sale of CO₂ compensation certificates, which is managed by the Swiss climate protection foundation, KliK. The sale of certificates is an essential aspect of the business case as the revenue from the biogenic CO₂ sales alone is not sufficient to cover production costs.

Before loading, the quality of the liquid CO₂ is tested. The monitoring and control of the food grade quality of the liquid CO₂ requires rather expensive equipment.

The CO₂ liquification plant was built by HZI (Hitachi Zosen INOVA) and was commissioned at the beginning of 2023. The project did start with a first feasibility study in 2019, construction was started in 2022. The investment was around 3 million CHF (3.5 million USD). The plant requires an area of approximately 80 m². It is estimated that personnel time for supervision and operation is 20 h/week. Plant maintenance will require an estimated downtime of 2 weeks per year. Further information can be found at Lehman et. al. 2024.



Figure 25: Impressions CO₂ Plant in Nesselbach

10 Conclusions

Methane is an important greenhouse gas and, due to its high global warming potential, the oxidation of methane is a cost-efficient emission reduction measure. Methane oxidation is a highly exothermic reaction and methane is valued as important energy carrier in form of natural gas. However, oxidation of gases containing low concentrations of methane, such as methane slips from CHP units or biogas upgraders, and older landfills, requires more sophisticated technologies.

The report reviews several technologies available to oxidize methane containing gases efficiently. Almost all of them allow a utilization of excess heat. In general, the lower the methane concentration in the fuel gas, the more effort is needed to minimise heat losses in order to reach the required temperature for a complete oxidation. Gases with methane concentration down to 0.37 vol. % can be treated in combustion systems without the addition of heat. Technically there are almost no limitations for emission reduction of lean gases. The abatement costs for such systems are in the range of related sectors as has been shown in the exemplar for RTOs. However, without support schemes for GHG emission reduction or taxes on GHG emissions, the implementation of such systems is more expensive than just emitting the methane, and consequently these systems are not competitive.

The suitable technology for the oxidation of methane is to be chosen based on several criteria:

- Composition of lean gas
 - Methane concentration
 - Contaminants (e.g. H₂S, siloxanes) - type and concentration
 - Oxygen concentration
- Methane volume flow (Capacity of the plant)
- Variations of flow and concentration (short and long term)
- Costs
- Demand for heat/energy on site

As described in the report there are many technical options available to oxidize methane. It can be oxidized without addition of energy to the combustion process down to concentrations as low as 0.37 vol. %.

Due to the high global warming potential of methane, oxidation is a rather cost-efficient emission reduction measure, even when more complex technologies such as an RTO are used. The technologies presented are state of the art and have many applications worldwide. They are ready for application and in most cases the infrastructure and services for implementation and operation are available.

Unlike thermochemical oxidation systems, biological systems do not have a guaranteed oxidation rate since its performance is depended on several parameters. These systems have shown effective methane reduction and greater adoption should result in more detailed information on performance and impact of design and operational parameters on the performance.

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