

Dissolved gas separation for engineered anaerobic wastewater systems

S. Heile¹, C.A.L. Chernicharo², E. M. F.Brandt³, E.J. McAdam^{1,*}

¹Cranfield Water Science Institute, Cranfield University, Bedfordshire, MK43 0AL, UK

²Department of Sanitary and Environmental Engineering, Universidade Federal de Minas Gerais, Belo Horizonte – MG, Brazil

³Department of Sanitary and Environmental Engineering, Universidade Federal de Juiz de Fora, Juiz de Fora – MG, Brazil

*Corresponding author: e.mcadam@cranfield.ac.uk

Abstract

Dissolved gases produced within engineered anaerobic processes subsequently create a fugitive emission which can have financial, environmental and health and safety implications. Whilst desorption technology has been used to control dissolved gases in the drinking water sector, there is considerably less understanding of its deployment in wastewater for which there are numerous existing and emerging challenges. This review therefore focuses on existing and proposed technological approaches to gas desorption in engineered anaerobic wastewater processes, with specific emphasis on technology compatibility and downstream gas phase management. Simplified engineered solutions such as diffused aeration and multi-tray aerators appear robust solutions for implementation into wastewater. However, these processes are characterised by a low mass transfer coefficient and require high gas to liquid ratios (G/L) to achieve reasonable separation, which suggests their suitability is limited to small scale applications, in which gas recovery is not a priority. Packed columns and membrane contactors afford process intensification through increasing interfacial area which favours large scale applications; although both will require prefiltration technology to obviate media clogging. Vacuum or steam is the preferred driving force for separation when gas recovery is sought, while sweep-gas is energetically favoured. Sweep-gas has been used for gas recovery by operating at G/L toward the equilibrium value, which somewhat constrains mass transfer. Process selection must therefore be weighted on whole life cost, but will also be dependent upon process scale, financial (e.g. incentivisation) and non-financial (e.g. carbon) instruments, which are strongly influenced by regional policy.

Keywords: *stripping, degassing, degasification; desorption, effluent, leachate*

1. Introduction

Desorption or stripping technology facilitates contact between water and gas in order to permit the separation of dissolved gases or volatile compounds from the liquid phase into the gas phase. Air stripping technology has been conventionally used to separate toxic organic compounds from drinking water for the protection of human health. Common application examples include the removal of trihalomethane (THM) compounds (Bilello and Singley, 1986) or the removal of light hydrocarbons (Ball and Edwards, 1992). Numerous commercial technologies are now available for the treatment of contaminated surface water, contaminated groundwater (Kavanaugh and Trussell, 1980) and for inclusion within the potable water supply network to provide treatment during distribution (Hirschhorn and Moore, 2014).

In contrast, there has been comparatively less attention paid to the application of desorption technology for wastewater treatment, for which there are numerous existing and emerging challenges that demand the separation of dissolved gases. Many of these opportunities relate to engineered anaerobic environments, where the presence of dissolved ammonia (NH_3), carbon dioxide (CO_2), hydrogen sulphide (H_2S) or methane (CH_4) gas pose operational issues (Table 1). In the UK, whilst a standard for dissolved methane has not been explicitly published by the Health and Safety Executive (the regulator), there is an expectation that dissolved methane in leachate is controlled before discharge into the public sewer system. Consequently, regulators in the UK and in many other countries conform to mine safety procedures (Last, 2017). The compliance limit is set by determining the dissolved methane concentration equivalent to the Lower Explosive Limit in the gas phase (LEL 5%) at 15 °C and 101.325 kPa ($1.4 \text{ mgCH}_4 \text{ l}^{-1}$), and applying a factor of safety of ten ($0.14 \text{ mg CH}_4 \text{ l}^{-1}$), and is widely adopted by most receiving sewerage authorities (Robinson and Carville, 2010; Cookney et al., 2016; Last, 2017). Air stripping is the standard methodology employed, with the air flow specified to ensure that the methane concentration in the waste gas is also sufficiently below the LEL to permit safe venting (Robinson and Carville, 2010). A key emerging area for engineered anaerobic technology is for full flow municipal wastewater treatment (Martin-Garcia et al., 2011) as the reduction in organic load in the absence of aeration coupled with the organic conversion to methane, suggest that energy neutral wastewater treatment is attainable even at ambient temperatures (McAdam et al., 2011). However, considerable dissolved methane losses have been reported due to the significant hydraulic flow and dilute organic concentration of municipal wastewater (Souza et al., 2011), which necessitates stripping to either limit carbon emissions (Brandt et al., 2016) or to recover methane to augment energy production (Mcleod et al., 2016). Air stripping has also been proposed for ammonia separation from sewage sludge either to reduce the energy demand for downstream processing of municipal wastewater

(Teichgräber et al., 1994) or to limit the effects of ammonia inhibition during anaerobic digestion, which is becoming increasingly evident with the advent of new feedstocks (Yenigun and Demirel, 2013).

In order to establish appropriate design criteria for desorption technologies applied to engineered anaerobic wastewater treatment systems it is possible that some knowledge transfer can be provided from desorption technology that has been applied to natural anaerobic environments. For example, most groundwater sources comprise a dissolved methane (CH_4) residual of *biogenic* origin, typically ranging $<0.01 \text{ mg l}^{-1}$ to 1.5 mg l^{-1} (Stolper et al., 2015; Darling and Goody, 2006). Scherer and Wichmann (2000) identified that air stripping was needed to limit methane concentration to 0.2 mg l^{-1} onto rapid gravity sand filters, to avoid blocking by large quantities of jellylike biomass produced as a result of biological methane oxidation. Air stripping has also been employed to reduce hydrogen sulphide (H_2S) in groundwater, as this is a common source of taste and odour complaint from customers; a H_2S residual of $0.05 \text{ mgH}_2\text{S l}^{-1}$ being sufficient for detection as an odour (Duranceau and Faborode, 2012). Whilst studies arising from natural anaerobic systems are insightful, similar to conventional potable air stripping studies, the utility of the arising knowledge in the design of wastewater desorption systems is somewhat constrained as wastewater composition is inherently more complex and the broad range of objectives, impose a diverse set of technology requirements which will demand very different operational boundary conditions. Furthermore, desorption technologies are differentiated by the method in which interfacial area is developed to initiate gas-liquid contact, through incorporation of packing media, the production of fine droplets, or the development of small gas bubbles, which will inevitably present new integration challenges when considering technological development for wastewater. Recent reviews have focussed on quantifying dissolved gas emissions (Mosse, 2013; Crone et al., 2017), whilst providing a précis on discrete gas-liquid separation technology (Crone et al., 2017), or contextualising potential solutions for a specific wastewater treatment plant (Mosse, 2013). This review complements and builds upon these previous studies, by critically examining existing and proposed technological approaches to gas desorption in engineered anaerobic wastewater processes, with specific focus on technology compatibility and downstream gas phase management, which has received little attention to date.

2. Water Chemistry

In a gas-liquid system, gas will partition into both the gas and liquid phase until equilibrium between both phases is reached. The extent of distribution between the two phases can be described by the partitioning coefficient or Henry's law constant. An increase in temperature increases water vapour pressure, such that the escaping water molecules at the gas-liquid

interface force other gas molecules away (Kemmer, 1987), which introduces a temperature dependency that can be described by (Sander, 1999):

$$H_i(T) = H_i^\circ \exp \left[-\frac{\Delta_{soln}H}{R} \left(\frac{1}{T} - \frac{1}{T^\circ} \right) \right] \quad (\text{Eq. 1})$$

where H_i° and T° are the Henry's law constant and temperature at standard conditions (T 298.15 K), $\Delta_{soln}H$ is the enthalpy of solution and R is the gas constant. The temperature dependence is provided by:

$$-\frac{d \ln H(T)}{d \left(\frac{1}{T} \right)} = \frac{\Delta_{soln}H}{R} \quad (\text{Eq. 2})$$

from which the gas-liquid partitioning can be determined at suitable reference temperature (Table 2). Gases arising from anaerobic conditions become increasingly soluble following a reduction in water temperature (Figures 1 and 2). Dissolved methane emissions are therefore particularly problematic from engineered anaerobic systems used in municipal wastewater applications, which are operated at ambient temperatures and are often characterised by substantial flow rates. To illustrate, between 45 and 85% of the total methane produced from an anaerobic reactor, has been demonstrated to emanate as an effluent emission (Cookney et al., 2016). A detailed description of Henry's law constants and temperature dependence coefficients is provided in Sander (1999). Since the Henry's constant represents gas-liquid equilibrium, this can be employed to estimate the volumetric gas to liquid ratio needed to achieve a prescribed removal rate, for desorption systems operated in sweep gas mode:

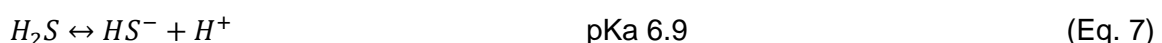
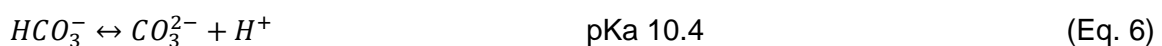
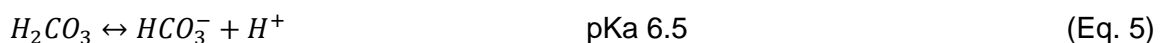
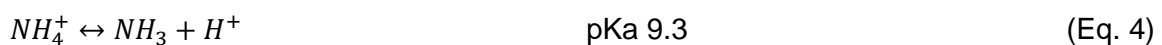
$$\left(\frac{A}{W} \right)_{equilibrium} = \frac{(C_0 - C_e)}{HC_0} \quad (\text{Eq. 3})$$

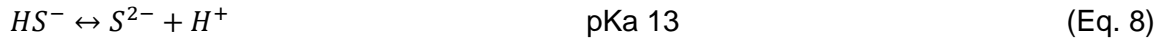
where H is the dimensionless form of the Henry's constant (Figure 1), and C_0 and C_e are the inlet and outlet concentrations respectively. This represents the minimum air to water ratio needed to achieve the prescribed treatment objective. The stripping factor (S) is representative of the separation potential:

$$S = \frac{A}{W} H$$

which implies that a stripping factor of one is sufficient for separation. In practice, a stripping factor of three is recommended to achieve the required treatment objective (Huang and Shang, 2006).

Such estimations for ammonia (NH_3 , Eq.4), carbon dioxide (CO_2 , Eq.5-6) and hydrogen sulphide (H_2S , Eq.7-8) are more complex since these solutes undergo ionisation, the extent of which is strongly associated with the pH of the water:





The dissolved concentration of a gas solute (c_i) can be estimated from the partial pressure (P_i) of the gas phase (Ho and Sirkar, 1992; Perry and Green, 2008):

$$c_i = H_i * p_i \quad (\text{Eq. 9})$$

The partial pressure of a gas (p_i) in the gas phase can be estimated based on the concentration dependency (mole fraction, y_i) and the total gas phase pressure (p_t):

$$p_i = y_i * p_t \quad (\text{Eq. 10})$$

Pauss et al. (1990) suggested that whilst the equilibrium concentration for highly soluble gases may be predictable, for poorly soluble gases, such as methane, the mass transfer limitation in the liquid phase could lead to supersaturation to as much as 80 times the thermodynamic equilibrium in anaerobic processes (Pauss et al., 1990). Methane supersaturation in effluent arising from anaerobic reactors has now been evidenced by numerous authors (Hartley and Lant, 2006; Souza et al., 2011; Cookney et al., 2016). Two mechanisms are generally proposed: (i) supersaturation arising from temperature and pressure transients, leading to an unsteady-state concentration; and (ii) the formation of microbubbles which entrain into the effluent (Hartley and Lant, 2006; Cookney et al., 2016). Pauss et al. (1990) proposed that in anaerobic processes, bubble nucleation emanates at the solid-liquid interface which is mediated at the surface of granular biomass or microscopic solid particles, which suggests that gas hold-up and entrainment into the effluent is possible. In a comparative study of an upflow anaerobic sludge blanket (UASB) reactor and membrane bioreactor (MBR) for municipal wastewater treatment, Cookney et al. (2016) noted that whilst the UASB effluent was supersaturated with methane, the MBR which comprised an ultrafiltration membrane for solids separation, the permeate dissolved methane concentration was at equilibrium, which tentatively supports the assertion that it is microbubble entrainment within the solids fraction that induces supersaturation (Hartley and Lant, 2006).

When the liquid or gas phase concentration is below the value predicted by Henry's law, there will be a transfer of mass between phases until equilibrium is reached (Govind, 2005). This can be practically achieved by diminishing either the mole fraction (y_i) or total pressure (p_t) of the gas phase by using sweep gas or vacuum as the driving force respectively (Eq. 10), to promote mass transfer. The rate of mass transfer is generally described by film theory, which assumes a stagnant film exists adjacent to the interface (Figure 3). As the fluid is considered stagnant, film transport is dominated by natural diffusion, thus Fick's law can describe mass transport across the film:

$$J = -D \frac{\Delta C}{\Delta X} \quad (\text{Eq. 11})$$

where ΔC is the concentration difference between the bulk solution and interface, and ΔX is film thickness. Fick's law evidences that to maximise mass transfer: (i) the maximum difference in concentration gradient between the bulk and interface should be maintained; and (ii) the film thickness at the gas-liquid interface should be kept as thin as possible. Whilst powerful, this analysis proposes that only a single stagnant film constrains mass transfer, whereas it is more generally proposed that stagnant films arise in both gas and liquid phases (two-film model). In general, for desorption processes focussed on separation of sparingly soluble compounds such as methane (a small Henry's constant), the liquid phase resistance dominates mass transfer (Ho and Sirkar, 1992). In contrast, for extremely soluble gases such as ammonia and hydrogen sulphide, it is the gas phase resistance which controls mass transfer (Yang and Cussler, 1986; Dvorak et al., 1996). To illustrate, for the separation of ammonia, a very soluble compound, high stripping air ratios of around $1497 \text{ m}^3_{\text{air}} \text{ m}^{-3}_{\text{H}_2\text{O}}$ are required for ammonia from sewage sludge (at high pH) to overcome the gas phase resistance (Kemmer, 1987).

3. Potential treatment technologies for gas desorption from anaerobic waters

3.1 Spray aeration

Spray aeration is simple to retrofit and possesses a small footprint (Figure 4a). These advantages have seen implementation in reservoir applications (Borkosky, 2014) and elevated storage tanks within potable distribution networks for THM removal (Hirschhorn and Moore, 2014). Water comprising dissolved gases is pumped through a manifold for distribution into small nozzles that induce droplet formation to create significant specific surface area. Sprinklers for droplet formation can be generally described as impact sprinklers or sprayheads. Single nozzle impact sprinklers employ fine nozzle sizes (around 3 mm) requiring considerable feed pressures (several hundred kPa) but produce droplets of median size (d_{50}) around 1 mm, which provides substantial specific interfacial surface area ($5998 \text{ m}^2 \text{ m}^{-3}$ assuming a spherical droplet). However, mass transfer is also dependent on drop height, droplet velocity and nozzle orientation (Kincaid et al., 1996) (typically oriented upwards). Glória et al. (2016) employed a free fall jet tower (Figure 4b) for simultaneous desorption of CH_4 and H_2S from a UASB treating municipal wastewater. Due to the droplets emerging from the free fall jet, the mass transfer mechanisms of the free draining tower tested by Glória et al. (2016) may be similar to the spray aeration, though less effective. The authors noted an average dissolved methane removal efficiency of 73% when employing a drop height of 1.1m compared to 62-63% at 0.5m, which demonstrates the important role of gas-liquid contact time (Table 3). To reduce feed pressure requirements, sprayheads employ coarser nozzle sizes (around 3 to 9 mm) to form a jet which impinges on a fixed or moving

deflector plate, to form d_{50} droplet sizes of around 0.7 to 2.53mm ($2359 \text{ m}^2 \text{ m}^{-3}$ assuming a spherical droplet) (Kincaid et al., 1996).

In drinking water distribution, the feed water pressure can be sufficient to compensate for the nozzle pressure drop. In comparison, engineered anaerobic processes are often constrained by lower pressure head availability. Furthermore, an increased particulate load from conventional anaerobic configurations (e.g. UASB) will constrain nozzle aperture selection. For example, the orifice size selected for municipal wastewater trickling filter arms is around 19 to 40mm in diameter to ensure they are free draining and low fouling (Ewida et al., 2006). Glória et al. (2016) implemented a 'free draining' spray tower design (free fall jet tower) and observed enhanced dissolved methane separation at a hydraulic loading of $0.132 \text{ m}^3 \text{ m}^{-2} \text{ min}^{-1}$ compared with earlier studies at considerably lower loadings but employing the same design (Souza et al., 2011). The enhancement can be ascribed to the increased entrance velocity inducing turbulence, and subsequent breakage to form a spray comprised of smaller droplet thus increase the available interfacial area. However, the considerably larger droplet sizes arising from free-draining spray systems will lower trajectories, interfacial area and, gas-liquid contact time due to the greater kinetic energy of the larger droplet sizes (Kohl et al., 1974; Kincaid et al., 1996).

Spray aeration is generally conceived to be efficient and notably can be coupled with natural ventilation (low energy requirement), although recirculation is sometimes demanded to achieve the specified treatment level. Nozzles can be fitted in downward configuration when used in conjunction with counter-current forced aeration to enhance separation (Stocking, 1988). In the simultaneous separation of CH_4 and H_2S using counter-current forced aeration, a reduction in sweep gas flow from G/L 1.59 to 0.77 did not impact upon dissolved methane removal, which is evidence that with an oversupply of air (S_{factor} 21-45), mass transfer is controlled by the liquid phase, which could be expected from the small Henry's constant (Ho and Sirkar, 1992; Glória et al., 2016). However, a reduction in sulphide removal from 80% to 57% was observed with the same G/L reduction, which implies that simultaneous removal of both compounds may be difficult to achieve for two compounds with such different Henry's law constants, once operating conditions have been optimised. Merrington and Richardson (1947) have shown that water near the periphery of the jet will result in small droplets while water near the core of the jet with the lowest relative velocity to the air will produce large droplets. Both interfacial area and contact time are therefore difficult to predict, which makes process design toward a specific treatment objective difficult. Since droplet size is dependent upon water velocity (Kincaid et al., 1996), such inconsistent effects may well be exacerbated in wastewater systems due to the transient flow rates experienced (Table 4).

3.2 Packed columns

For low loading rates, drip plates can be used to distribute water at low hydraulic loading rates whereas for high load rates, spray nozzles are preferred to promote uniform distribution onto the packing media at the top of the column (Vinci et al., 2006). Packing typically comprises of structured media that promotes high specific surface areas of around $250\text{--}300\text{ m}^2\text{ m}^{-3}$ (Orlando et al., 2009). The random packing media allows development of a thin water film, with substantial specific interfacial surface area and also provides some interstitial mixing which augments gas-liquid contact (Figure 4c). Vinci et al. (2006) suggest operational hydraulic loading for CO_2 stripping was between $36\text{ to }108\text{ m}^3\text{ m}^{-2}\text{ h}^{-1}$ dependent upon packing type, which are considerably higher than comparative technologies. Column diameter is therefore selected in accordance with the specific packing material to constrain flooding; a phenomenon where high up-flow gas velocities impose sufficient resistance to down-flowing water to restrict flow. Gas velocities are typically specified at 50% of their flooding value (Geankoplis, 2003). To eliminate corrosion problems, specifically pertaining to sulphide, fibre glass and wooden construction have been used (Kemmer et al., 1987), although other materials (e.g. polypropylene, stainless steel) are also applicable.

Air is typically used as the stripping gas to remove dissolved gases such as CO_2 , NH_3 , H_2S or CH_4 (Kemmer et al., 1987). Scherer and Wichmann (2000) tested several column configurations for methane desorption from groundwater, and identified 50-60 % removal efficiency with an empty bed contact time (EBCT) of 230s. The authors noted that increasing the G/L ratio from 5 to 30 did not influence mass transfer, and suggested an increase in EBCT was required to improve removal. Packed columns were preferred when over 90% methane desorption was required, which can be enabled through increasing column height above that employed in their study (h , 0.6 m). A vacuum driven column packed with pall rings was supposed to be constructed in the Netherlands to treat supersaturated groundwater for dissolved methane, which was expected to recover 90-95% of methane operating at 5kPa (after Mosse, 2013). The disadvantage of vacuum desorption is the comparatively high energy requirement (Scherer and Wichmann, 1987). Through reduction in packing fraction, it is possible to achieve pressure drops below 2.4 to 7.5 mbar for air stripping (Kemmer et al., 1997; Crittenden et al., 2005). Such low pressure drops enable specification of fans with a low pressure ratio, rather than more costly blowers (with high pressure ratio), which enables economic desorption. Interestingly, at G/L ratios below 5, Scherer and Wichmann (2000) noted enhanced mass transfer for methane versus CO_2 at low G/L ratios, whereas mass transfer was similar for both gases at higher G/L ratios, which requires consideration when simultaneous separation is preferred.

Steam stripping has been applied for sour condensates which contain both ammonia and sulphide. As both ammonia and H_2S are ionised, considerable steam is required to

overcome the lower vapour pressure, than would be exhibited in the correct pH range. Nevertheless, 87% and 100% removal of ammonia and sulphide was respectively demonstrated from a sour water (feed 1480 and 1876 mg l⁻¹) at pH 9 using a column packed with Raschig rings (Kemmer et al., 1987). When waste heat is available, thermally driven stripping may be more economically appropriate than chemically induced stripping. For example, thermal ammonia stripping from landfill leachate has been operated on the waste heat from a biogas combined heat and power system (CHP) in which the temperature is raised to 65 to 70°C, requiring around 1MW heat to treat 80-100 m³ d⁻¹ leachate (Organics, 2017b).

For methane desorption from natural anaerobic systems, pretreatment (deferrisation) was recommended to limit iron precipitation in the column (Scherer and Wichmann, 2000). During ammonia stripping from landfill leachate, carbonate precipitation has similarly been observed (Organics Ltd., 2017a). For full flow anaerobic applications, the precipitation of elemental sulphur is also feasible when sweep gas is applied:



Due to the low oxidation potential of oxygen, the precipitation of sulphur is more probable for technologies with extended residence times (McVay, 2011; Filtronics, 2017). Furthermore, whilst the stoichiometric oxygen demand for precipitation is around 0.5gO₂ gH₂S⁻¹, in practice, the oxygen demand is around five times higher to force the reaction (McVay, 2011). For illustration, if operation is fixed to the minimum G/L ratio for dissolved methane removal (~0.032) and around 20% oxygen transfer efficiency is assumed, then a dissolved oxygen residual of only 1.9 mgO₂ l⁻¹ is achieved, which suggests that it is gas-liquid systems operating in excess of the minimum G/L ratio would incur the greatest potential for precipitation. This is likely to demand increased maintenance coupled with the potential need for downstream separation of insoluble colloidal sulphur (McVay, 2011). In addition to inorganic fouling, organic fouling and clogging can also be expected for engineered anaerobic systems. Packed columns are difficult to clean once blocked due to restricted access to media (Kemmer et al., 1987; Stocking, 1988). As such, pre-treatment is likely to be demanded. However, there is only limited information on pretreatment in the literature. Whilst further work is required to ascertain pretreatment design, commercial scale ammonia air strippers for return liquor treatment (comprised of a higher solids and organics fraction than dilute wastewater) suggest 2mm diameter coarse filtration is sufficient to maintain operability (Organics Ltd., 2017a; Organics Ltd., 2017b).

3.3 Tray aerators

Tray aerators are commonly used for natural anaerobic systems and have been applied for the simultaneous separation of CO₂ and H₂S (Duranceau et al, 1999; Lochrane, 1979;

Duranceau and Faborode, 2012). Feedwater is distributed from the manifold into the top tray, subsequently flowing under gravity over the sides into trays below, eventually entering into an open collection basin at the base (Figure 4d). Even distribution of water over each tray is essential for effective sulfide treatment (Duranceau and Faborode, 2012). Trays can also be filled with media to increase interfacial area and encourage mixing (Kemmer et al., 1987). Trays are typically spaced between 0.3 and 0.75m apart (Duranceau and Faborode, 2012) which reduces pressure drop to allow natural ventilation as the sweep gas. This construction enables easy access to trays for cleaning (Kemmer et al., 1987), and will limit clogging, which are important considerations for wastewater applications. Wells (1954) identified 35 to 45% sulphide removal using a multiple tray aerator which is supported by full scale survey data of multiple tray aerators comprised of four tray stages, each stage comprising up to six trays (Duranceau and Faborode, 2012).

Low-profile or sieve tray aerators introduce forced draft aeration underneath perforated trays to develop an air-water foam with high specific area and mixing. An increase in the air flow to positive-draft aerators can increase the effectiveness of the addition of oxygen to water, or the removal of H_2S , compared with normal tray aerators (Duranceau and Faborode, 2012). Process height is constrained as efficiency is more closely governed by the length and width of the trays than overall process height (Wessels, 2014). This is potentially advantageous for incorporation downstream of engineered anaerobic processes to avoid the requirement for intermediate pumping. However, 'low-profile' aerators often require up to twenty times the air flows compared to packed bed systems (Mead and Leibbert, 1998).

3.4 *Diffused aeration*

Full scale diffused aeration schemes have been constructed for both natural (Schippers and Schotsman, 2010; Mosse, 2013) and engineered anaerobic environments (Robinson and Carville, 2010). Submerged aerators are used to produce small diameter bubbles that provide substantial air-water interfacial area (Figure 4e). However, a combination of short bubble residence times and resistance to mass transfer demand large air/water ratios to achieve analogous removal efficiencies to packed columns (Bilello and Singley, 1986). Mass transfer can be improved by using plate aeration, which has been used in the Netherlands for dissolved methane removal from groundwater (Schippers and Schotsman, 2010; Mosse, 2013). Most diffused aeration systems employ diffusers sited on the base of a tank which exerts a high gas pressure headloss, although when deployed in open tanks the water pressure losses are comparatively negligible. Khan et al. (2011) developed an aeration-settling system comprised of fine pore submerged diffusers to treat UASB effluent. Whilst aeration was implemented for the biological oxidation of organics, the authors noted

simultaneous H₂S stripping despite short residence times (30 minutes). When high dissolved gas removal efficiencies are required, diffused aeration is generally not recommended (Kavanaugh and Trussell, 1981 after Crittenden et al., 2005). However, Robinson and Carville (2010) demonstrated a diffused aeration design to achieve >99% dissolved methane removal efficiency from landfill leachate. The authors proposed cascade aeration comprised of four tiers, which drained sequentially by gravity to each subsequent tier, each tier comprising of an independent air supply. Diffused aeration was chosen for this application as the robustness of this technology is proven for wastewater implementation. However, high G/L ratios (S_{factor} 280-420) and long retention times (180 minutes) suggest the technology is best suited to small scale applications not seeking to recover the dissolved gases for reuse (Table 5).

3.5 *Membrane contactor*

Membrane contactors (Figure 4f) are commercially mature for high value gas-liquid applications and are increasingly replacing conventional gas-liquid technology for a broad range of commercial desorption applications (Wang et al., 2017). The hydrophobic microporous membrane facilitates non-dispersive contact between gas and liquid phases, leaving the gases free to diffuse through the open pores (Heile et al., 2014). Through this phase separation, traditional technology challenges such as foaming and flooding are avoided which enable surface loading rates over an order of magnitude higher than conventional systems (Hofkes et al., 1983; Gabelman and Hwang, 1999). The membrane introduces a third mass transfer resistance (Figure 3), although this is generally negligible when the membrane is microporous, thus the mass transfer coefficient is similar to packed column technology (Bandini et al., 1992; Belaisaoui et al., 2016). It is the increased specific surface area that enables a ten-fold increase in process intensification versus packed columns (Belaisaoui et al., 2016). Due to phase separation, pressure drop is independent of phase mixing effects and is instead controlled by the interstitial spacing which present reasonably low pressure drops, and since the technology is enclosed, the hydraulic head is not broken which can be energetically advantageous for engineered anaerobic systems (McAdam, 2015).

Membrane contactors have not seen commercial deployment in anaerobic systems generally but have been the most widely explored technology for dissolved methane recovery from engineered anaerobic wastewater systems (Cookney et al., 2012, 2016; Mcleod et al., 2016; Henares et al., 2016) and their application to methane recovery from groundwater has also been considered (Wessels, 2014). Cookney et al. (2016) demonstrated up to 99% dissolved methane removal efficiency with retention times of between 1.5 and 12.5 s. It has been suggested that the significant interfacial area (around

4600 m² m⁻³) imposed through tight hollow fibre packing will permit the use of low G/L ratios for methane stripping, which will reduce energy requirements for recovery, and can be further improved through the use of transverse flow to improve dispersion through the fibre bed (Mcleod et al., 2016).

When introduced into complex wastewater, water quality and changes to surface properties due to fouling can induce pore wetting, which markedly reduces mass transfer (Heile et al., 2014). To circumvent such effects, nonporous membranes can be used in which mass transport is dependent upon the gas solutes solubility and diffusivity within the polymer, which will increase resistance to mass transfer (Heile et al., 2014). Bandara et al. (2011) used such a composite fibre which comprised nonporous polyethylene layered onto a porous polyurethane support for dissolved methane removal from anaerobic effluent produced with an UASB treating synthetic domestic wastewater. The authors successfully demonstrated this fibre construction using residence times up to 9.2 h in the degassing stage; the extended residency may have been necessary to compensate for the lower gas permeability across the nonporous membrane. Cookney et al. (2016) demonstrated that commercial microporous membrane contactors downstream of an anaerobic Membrane Bioreactor (MBR) was analogous to when operating on clean water which suggests negligible impact of residual organics in the permeate produced by the upstream 0.03µm pore size ultrafiltration membrane used in MBR. Due to the tight interstitial membrane fibre packing, upstream filtration may also be necessary to obviate bed clogging. Ten micron prefiltration is recommended although up to 40 microns has been successfully trialled (Henares et al., 2016). When operating a nonporous polydimethylsiloxane membrane contactor downstream of a UASB (and 40 micron stainless steel filter), Henares et al. (2017) demonstrated that surface fouling reduced productivity but fouling was mostly reversible through a combination of physical/chemical cleaning. To avoid prefiltration, Cookney et al. (2012, 2016) reduced fibre packing to 364 m² m⁻³ to promote channels of sufficient size to avoid clogging. Whilst successful, the reduction in interfacial area reduces the economic and operational competitiveness versus conventional technologies, and it is therefore proposed that membrane contactor systems are an attractive technology when coupled with upstream pre-filtration.

4. Gas phase management

4.1 Downstream gas phase management for discharge to the atmosphere

Carville and Robinson (2010) described the application of an excess sweep gas when using diffused aeration (Stripping ratio, around 400) for landfill leachate. The excess sweep gas provided three purposes: (i) providing good gas-liquid contact through agitation; (ii) ensuring the gas-phase did not constrain mass transfer in order to provide high treatment efficiencies

(~99%); and (iii) to ensure the off-gas methane concentration was below the lower explosive limit for safe venting (around 0.028% v/v compared to 5% v/v for the Lower Explosive Limit). Landfill leachate represents a small liquid discharge (typically between 50 and 3000 m³ d⁻¹) with dissolved methane concentrations ordinarily in the range of between 3 and 15 mg l⁻¹ (Marinheiro and Law, 2015). As such, the arising carbon emission is negligible and off-gas treatment is not ordinarily required for carbon abatement. However, since CH₄ is 25 times more potent as a greenhouse gas compared to CO₂, off-gas treatment must be considered to diminish arising carbon emissions in larger applications (IPCC, 2007). Whilst numerous research studies have been published on biofiltration for low methane content waste gas (250-50000 ppmv) in landfill, and agricultural applications, there are few operating on waste gas arising from engineered anaerobic systems for municipal applications (Melse and Vander Werf, 2005; Gebert and Gröngroft, 2006; Munoz et al., 2015). Biofiltration has been successfully demonstrated for the oxidation of methane in off-gas at low concentrations from 0.17%v/v to 3.63%v/v, requiring residence times of between 7.4 and 42.8 minutes (Brandt et al., 2016; Nikiema et al., 2005). Temperature has been determined to considerably influence oxidation rate (Menard et al., 2009). Between 100 and 500 ppmv H₂S has been measured in strip-gas (Glória et al., 2016). An emerging need is therefore to establish whether biofiltration systems that can mutually treat both hydrogen sulphide and methane simultaneously (Glória et al., 2016). Whilst not commercially mature, biofilters represent a reasonably low maintenance, cost effective option for point source carbon abatement, relative to the other technical options which may be more appropriate for higher off-gas methane compositions (Table 6). For dissolved ammonia concentrations exceeding 1000 mgNH₃ l⁻¹, off-gas treatment requires consideration, which can be achieved through thermal oxidation or gas scrubbing to precipitate as a salt (Organics Ltd., 2017a).

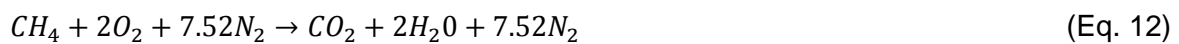
4.2 Downstream gas phase management for recovery and re-use

Due to the Henry's constant for methane (H_{CH_4} around 28), the theoretical G/L_{ratio} required to achieve reasonable dissolved methane removal is small and as such provided the gas phase can approach equilibrium (the underlying assumption of the theoretical gas to liquid ratio, Eq. 3), then modest methane compositions can be achieved in the sweep gas which could be sufficient for re-use. Mcleod et al. (2016) demonstrated this concept within a hollow-fibre membrane contactor for dissolved methane recovery from a saturated solution (Table 5). The authors showed that reducing the G/L_{ratio} enriched the gas phase, but the lower Q_g also constrained removal efficiency ostensibly through an increase in gas phase resistance. It is generally recognised that to achieve good single stage treatment, the operational G/L_{ratio} should be considerably above that of the equilibrium (Figure 5). To illustrate, experimental data for Chloroform desorption which compares diffused aeration to

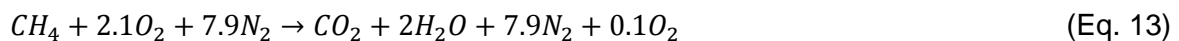
packed columns, has been reproduced and compared to the theoretical G/L_{ratio} (Bilello and Singley, 1986). Several important observations can be made: (i) Process selection (specific surface area, agitation, film thickness and contact time) strongly influences the extent of removal at equivalent operational G/L_{ratio} ; (ii) A reduction in operational G/L_{ratio} toward the theoretical G/L_{ratio} lowers single stage treatment capacity, which would suggest the onset of gas phase controlled mass transfer; (iii) The operational and theoretical G/L_{ratio} can intersect at lower G/L_{ratio} ; and, (iv) at the point of intersection, the corresponding removal efficiency is explicitly dependent on process characteristics (the packed column in this example achieving considerable higher removal efficiency at lower G/L_{ratio}). Matsuura et al. (2015) evidenced operation close to the equilibrium line when using a hanging sponge desorption column (Stripping factor between 1.2 and 1.8, Table 5). Despite operating close to the theoretical G/L_{ratio} , the authors achieved 70% dissolved methane removal efficiency and an off-gas methane composition of 39.4%. Scherer and Wichmann (2000) evidenced that the mass transfer coefficient for methane was 1.5 times higher than carbon dioxide at a G/L_{ratio} of 5, and was 3 times higher at a G/L_{ratio} around 1, which could further influence the resultant composition of the gas-phase.

Mcleod et al. (2016) also achieved reasonably concentrated methane in the off-gas using a hollow fibre membrane contactor and with much shorter residence times (300s versus 7200s). The authors suggested that dissolved methane removal efficiency could be further enhanced by improving shell-side dispersion through incorporation of transverse rather than parallel flow membrane module construction. Interestingly, in their study, higher methane composition was identified within the gas phase than was projected by mass balance. This enrichment was asserted to arise from counter-diffusion of the inert sweep-gas (principally oxygen and nitrogen) into the liquid phase, which had been previously modelled by Ahmed and Semmens (1992) following study of water oxygenation using hollow fibre membranes. Data from Matsuura et al. (2015) supports this position, as the authors were unable to detect Oxygen in the off-gas within a sweep-gas initially comprised of air. Importantly, this mechanism suggests it is possible to enable enrichment at operational G/L_{ratios} higher than the equilibrium, thus providing both treatment and recovery.

The specific methane composition required in the off-gas for combustion can be estimated based on combustion stoichiometry (Flagan and Seinfeld, 1988):



The stoichiometric air required to facilitate combustion is therefore around $10.4 \text{ m}^3 \text{ air m}^{-3} \text{ CH}_4$. In practice, combustion is achieved in the presence of a stoichiometric excess of air (e.g. 5% stoichiometric excess):



To illustrate the significance of the stoichiometric requirement of air to the methane composition in the recovered gas phase: 1m³ of recovered gas which comprises 25% CH₄ and is combusted in the presence of a 5% stoichiometric excess of air (2.725 m³) will be diluted pre-ignition to 6.7% CH₄. This is comparable to the volume fraction of methane employed for methane-air ignition (6.5 and 10.5% v/v; Zhang and Li, 2013) and is within the flammability limits for methane ignition (5 to 15% v/v). Whilst this example evidences that use of low CH₄ composition for ignition is achievable, commercial micro-turbines and larger scale CHP engines are ordinarily specified to operate at minimum CH₄ compositions of 30% and 40% respectively (Relea et al., 2009). In the case of anaerobic systems used for methane recovery, there will be two main recovery routes: methane arising from the headspace of the anaerobic reactor and dissolved methane from the effluent. The gas phase composition of the headspace methane for municipal applications ranges between 55 and 85% (Lester et al., 2013) dependent on climatic and operational conditions. As the methane mass flow arising from the anaerobic reactor headspace will be generally equal to or more considerable than the mass flow arising from the dissolved fraction (McAdam et al., 2011; Cookney et al., 2012), the final pre-ignition methane composition can be controlled through blending.

4.3 Alternative methods for gas phase concentration and re-use

An alternative approach to concentrating methane in the gas phase is to change the applied driving force (Figure 6). Vacuum has been employed at commercial scale in wastewater to improve sludge settling through the stripping of nitrogen gas. A vacuum of around 50 mbar is applied above an enclosed tank sited within a tower configuration. In this application, the specific remit is to circumvent nitrogen supersaturation, by removing micro bubbles which support sludge bulking. Whilst successfully demonstrated for this application, the pressure drop, mixing profile and interfacial area available for gas-liquid exchange in the present configuration may not be economically or operationally appropriate for anaerobic applications which are seeking to achieve very low dissolved residuals at minimum energy (Cookney et al., 2016). Sweep gas rather than vacuum is common place for the broader range of municipal stripping applications (water and wastewater), including the separation and recovery of ammonia which is conducted in a two-stage stripper-scrubber arrangement (Christian et al., 2016). However, the proposed construction of a vacuum driven (50 mbar) packed column comprised of pall rings was reported to be under construction for the recovery of methane from a supersaturated groundwater in the Netherlands (Schippers and Schotsman, 2010; Schotsman (2012) after Mosse, 2013). Several pilot scale studies using hollow fibre membrane contactors have also demonstrated use of vacuum for the separation of dissolved methane from the effluent of Upflow Anaerobic Sludge Blanket (UASB) reactors

treating municipal (Bandara et al., 2011; Cookney et al., 2012, 2016) and synthetic industrial wastewaters (Henares et al., 2016). Bandara et al. (2011) provided one of the only studies to quantify methane composition of the gas phase recovered under vacuum. In their study, a lower than expected gas phase CH₄ composition of 22% was recorded, which was asserted to arise from air ingress into the dissolved gas collection line.

Vallieres and Favre (2004) suggested that vacuum may be more favourable when gas phase purity is important as vacuum obviates dilution; however, the authors demonstrated that sweep gas is favoured from an energy perspective. In practice, the vacuum required for separation is specified to reduce partial pressure of the gas to less than 50% of the partial pressure at equilibrium with the desired effluent concentration, and is therefore analogous to operating sweep-gas with a stripping factor greater than one (Kemmer, 1987). The highest economic return from the recovered methane has been identified through a trade-off between capital cost and operational cost (Cookney et al., 2016). The capital cost of the driving force must therefore also be considered. For sweep gas mode, very low gas phase pressure drops can be achieved (less than 7 mbar) which favour specification of fans (rather than blowers) which offer high air flows, require little energy and cost considered less than vacuum technology. Whilst a definitive decision on the most appropriate driving force for recovery has yet to be made in the literature, as several authors have now identified gas phase enrichment using sweep gas, the whole life cost, and not simply the energy balance, may favour sweep gas. Steam stripping has been used to recover dissolved methane from sour gas to provide an enriched gas phase (Kemmer, 1987). Whilst heat recovery is achievable from CHP systems, the available heat is sufficient to support high flowrate engineered anaerobic systems. For example, using the CH₄ yield previously reported for both headspace and dissolved methane, and assuming 60% thermal conversion (McAdam et al., 2010), the available heat is around 0.41 kWh m⁻³ wastewater treated which is sufficient to raise fluid temperature by around 0.5°C. However, steam stripping of ammonia from anaerobic digestion return liquors (a comparatively small flow) has been considered viable using CHP waste heat, which it is asserted can achieve an aqueous ammonium hydroxide product of up to 20%, which offers a potential opportunity for re-use (Teichgräber and Stein, 1994).

5. Conclusions

From this review, several candidate technologies have been identified for gas desorption from engineered anaerobic environments. Both diffused aeration and multi-tray aerators appear robust to complex waters and are simple to maintain, making them good candidate technologies for application to wastewater. Multi-tray aerators also offer low pressure drops and limited process height, as removal is more dependent upon tray surface area, which is

advantageous to integration within wastewater flowsheets. However, high G/L ratios are required and the separation efficiency is comparatively poor. Packed columns and membrane contactors offer process intensification through incorporating high interfacial area, but are expected to require pre-treatment in order to ensure consistently robust performance over time; integration downstream of an MBR is an example of one synergistic opportunity. In establishing the value of process intensification, consideration should be given to the comparatively low G/L ratios and enhanced removal efficiencies that can be achieved. Regardless of the technology selected, construction materials are important and should be specified to avoid H₂S induced corrosion.

Vacuum is generally considered favourable for recovery and reuse of dissolved gases, however, sweep gas is energetically favourable as the driving force for separation. Several authors have now evidenced that sweep-gas mode can deliver sufficient gas-phase methane concentrations for re-use by reducing the G/L ratio toward G/L_{min} ($S_{factor} \sim 1$). Whilst this decreases single stage removal efficiency, it may present an economically attractive option for recovery. If further separation is needed, the reduction in efficiency can be compensated for through two-stage processing which has been advocated elsewhere (Matsuura et al., 2015). Operating membrane contactors in transverse flow is also expected to improve single-stage processing through enhanced dispersion. High separation has been demonstrated below G/L_{min} in transverse mode using combined sweep-vacuum.

Several studies have sought to achieve simultaneous gas desorption. However, marked differences in solubility and dissociation chemistry make this difficult to realise in practice. The decision for which technology to invest in and how best to manage the resultant gas phase will be primarily dependent upon process scale. It is proposed that for:

- Small scale liquid emissions (e.g. landfill) robustness will be favoured over energy savings, with no requirement for gas phase management;
- Medium scale liquid emissions, economic considerations are increasingly important and abatement technology will be applied for gas phase management;
- Large scale liquid emissions, dissolved methane recovery will be the primary focus to enable recovery and re-use.

The boundary between these process scales will depend upon the weighting applied by financial and non-financial instruments such as incentivisation for renewable energy and carbon accounting, which are strongly influenced upon local economies.

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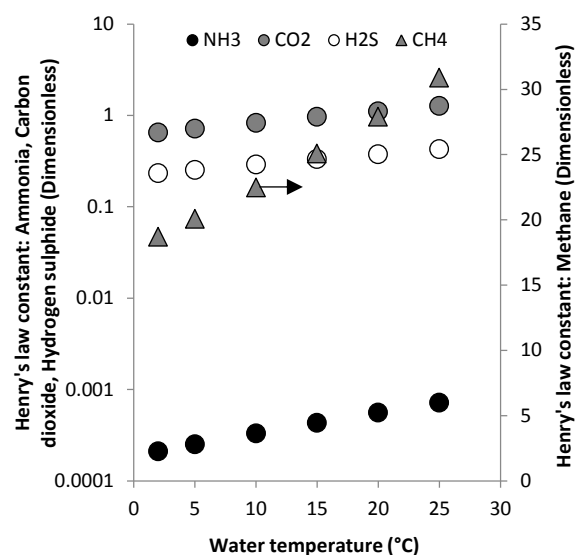


Figure 1. Comparison of dimensionless Henry's law constants for the common gases emanating from engineered anaerobic environments.

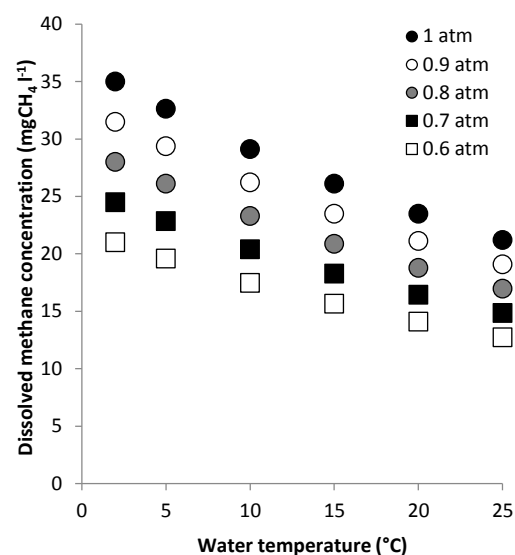


Figure 2. Demonstrating the influence of gas phase partial pressure and water temperature on dissolved methane concentration.

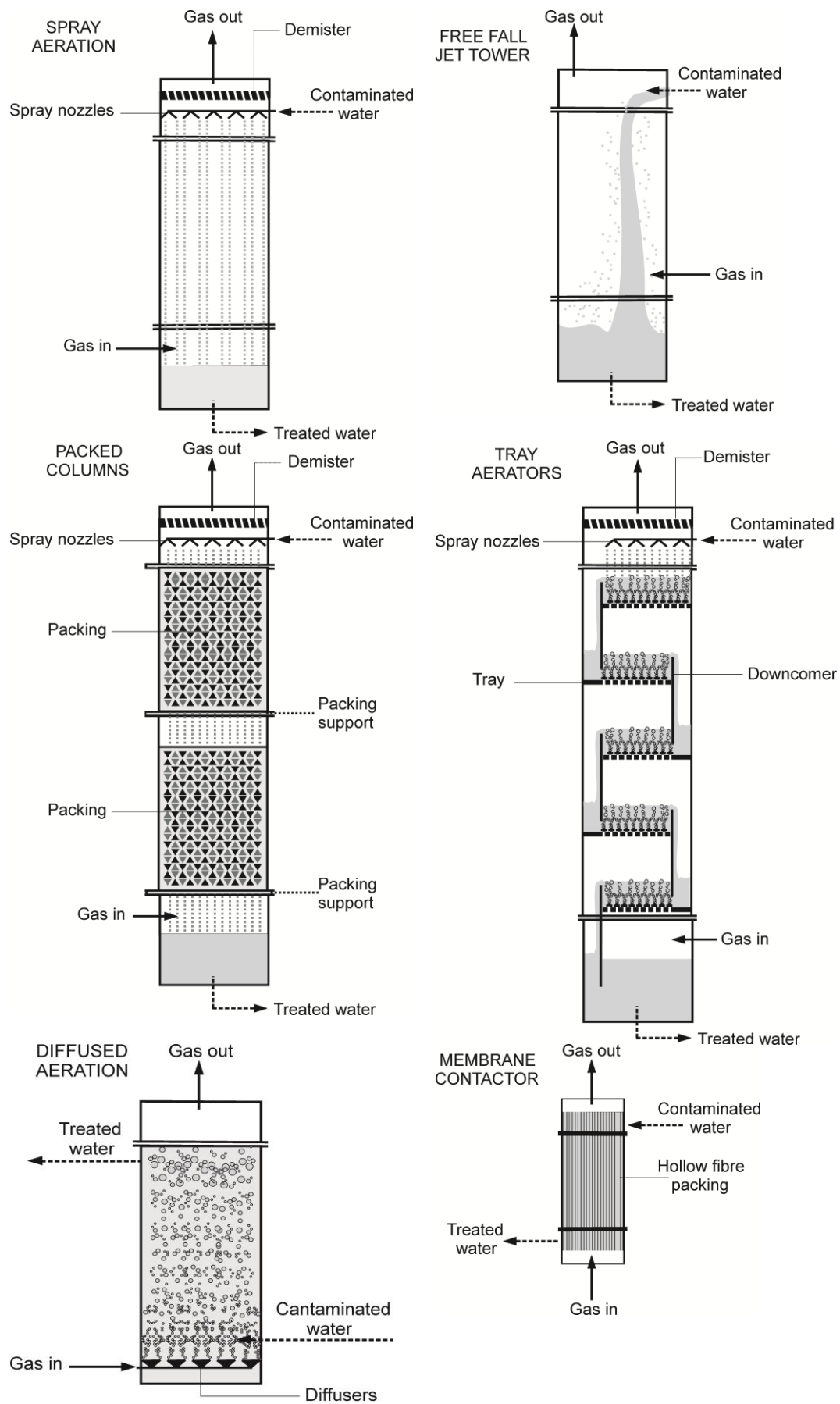


Figure 3. Schematic representation of the treatment technologies for gas desorption from anaerobic waters

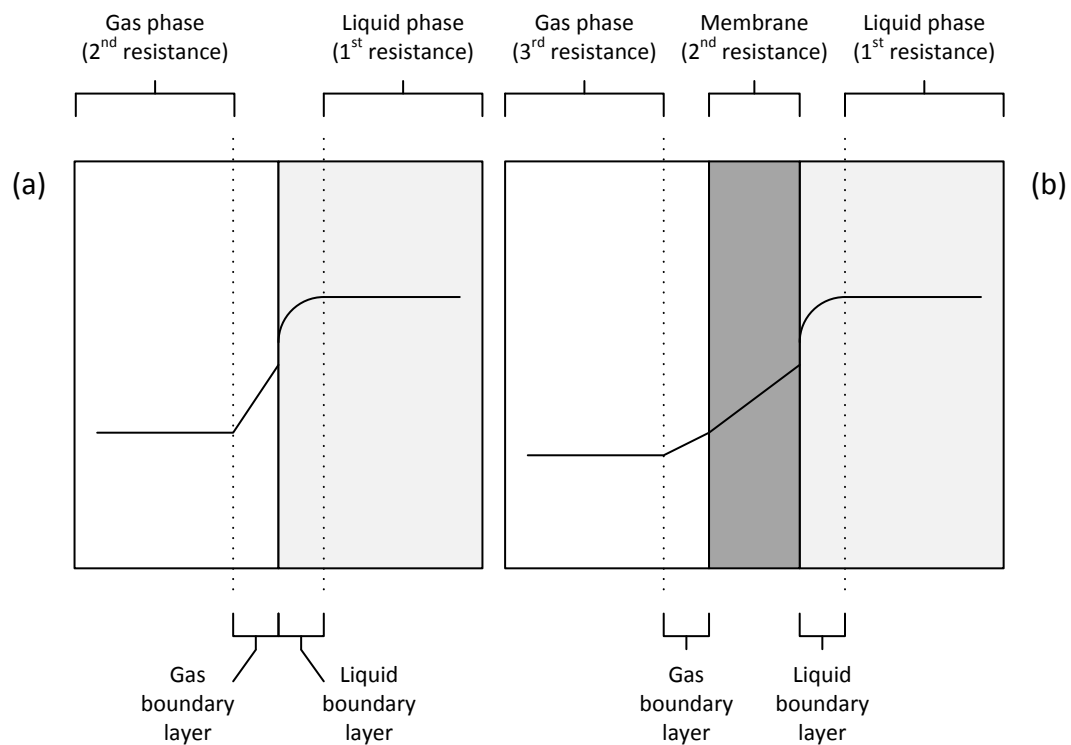


Figure 4. Schematic representation of two-film theory applied to conventional desorption technology (a); and membrane based desorption technology (b).

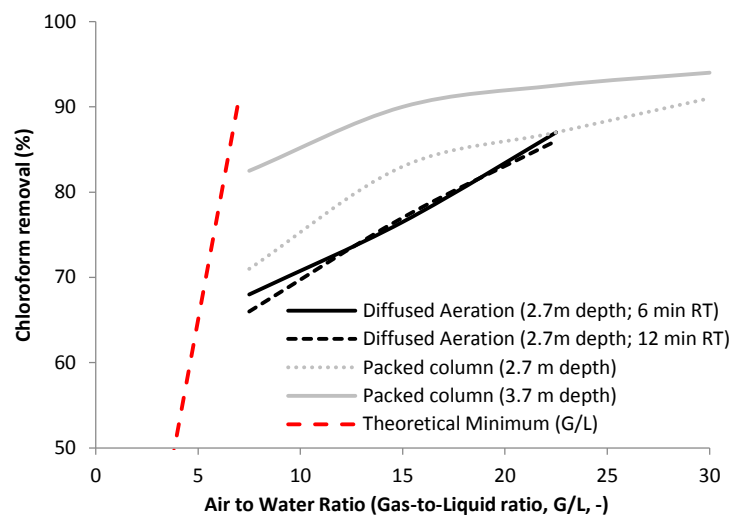


Figure 5. Desorption data demonstrating separation behaviour as G/L is reduced toward the minimum G/L (Eq. 3). Experimental data adapted from Bilello and Singley (1986).

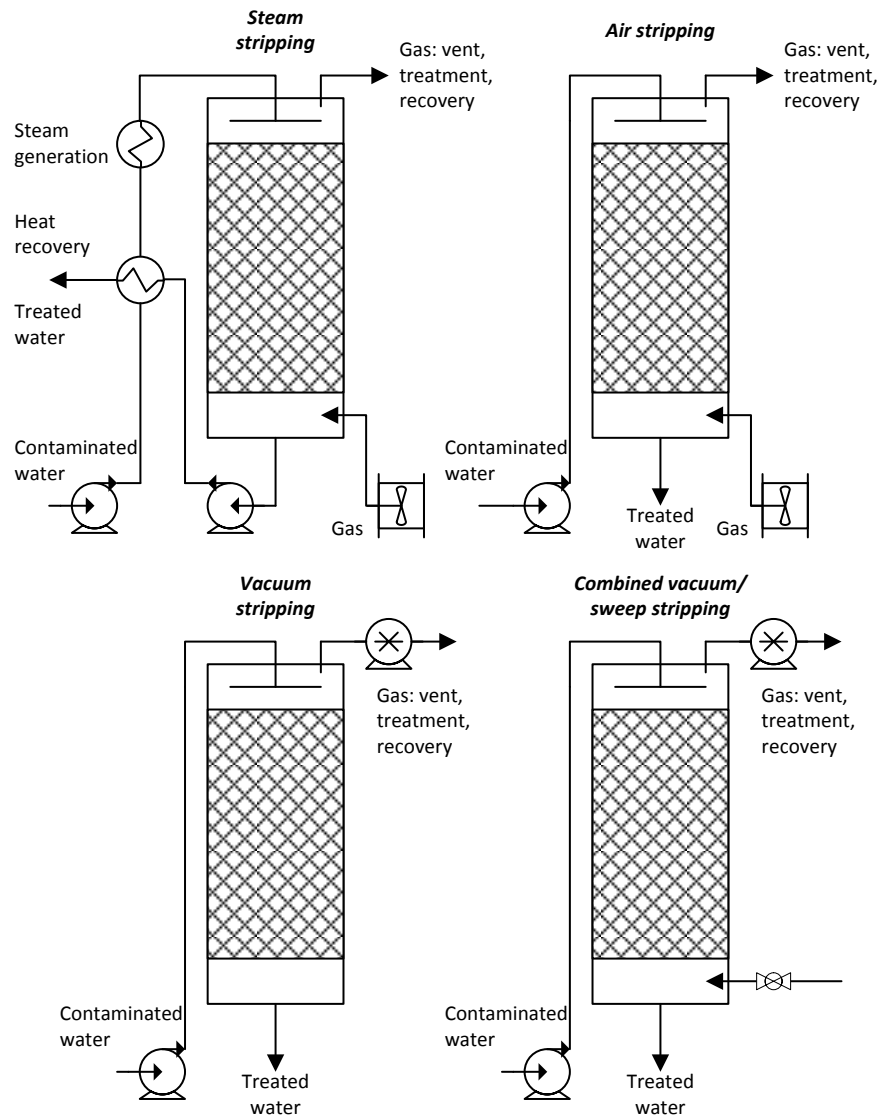


Figure 6. Example schematics of the four main driving forces employed for desorption: Steam, Air, Vacuum, Combined Sweep/Vacuum.

Table 1. Gases arising from anaerobic process that can be separated using desorption technology

Driver	Compound	Application/ Constraint	Guideline value/ Key performance indicator	References
Downstream treatment	NH ₃	Wastewater sludge/ Reduce energy downstream or limit NH ₃ inhibition in AD	Guideline value for inhibition varies, 2800 to 6000 mg l ⁻¹ TAN	Yenigun and Demirel, 2013
	CH ₄	Groundwater/ Filter blocking	<0.5 mg l ⁻¹ for deferrisation in rapid sand filter <0.2 mg l ⁻¹ for nitrification and demanganisation in rapid sand filter <0.2 mg l ⁻¹ no treatment required	Scherer & Wichmann, 2000
	H ₂ S	Groundwater & wastewater/ Reduce corrosion	Gas phase: 1ppmV, extensive damage to electrical infrastructure Liquid phase: 0.1-0.5 mgS l ⁻¹ minor corrosion; >2 mgS l ⁻¹ severe corrosion	USEPA, 1991; Zhang et al., 2008; Hvitved-Jacobsen et al., 2002
Product water quality	H ₂ S	Groundwater/ Taste and odour	<0.05 mg l ⁻¹ for low odour impact <1 mg l ⁻¹ 'swampy' odour >1 mg l ⁻¹ 'rotten egg' odour	Duranceau and Faborode, 2012
Carbon reduction	CH ₄	Municipal wastewater/ Carbon neutrality	^a 88% recovery	Mcleod et al., 2016
Health and safety	CH ₄	Landfill leachate/ Safe disposal to sewer	Gas phase: Below LEL (5%) ^b Liquid phase: <0.014 mg l ⁻¹	Robinson and Carville, 2010; Cookney et al., 2016
Energy recovery	CH ₄	Municipal wastewater/ Electrical power from CHP	85-90% recovery most economically efficient	Cookney et al., 2016

AD – Anaerobic Digestion. NH₃ – Ammonia; CH₄ – Methane; H₂S – Hydrogen sulphide; CHP – Combined Heat and Power; LEL – Lower Explosive Limit; TAN – Total ammonia nitrogen. ^aThis assumes a CO₂ offset of 0.543 kg kWh⁻¹ against grid produced electricity. ^bBased on liquid phase concentration equivalent to 5% methane in the gas phase at ambient pressure, combined with a safety factor of 10.

Table 2. Diffusion and partitioning coefficients. Partitioning parameter enable temperature correction of Henry's law constant (adapted from Sander, 1999).

	Partitioning coefficient ^c		Diffusion	
	H_i^0	$\frac{d \ln H(T)}{d \left(\frac{1}{T} \right)}$	D_{water}^d	D_{air}^e
	(mol dm ⁻³ atm ⁻¹)	(K)	(x10 ⁻⁹ m ² s ⁻¹)	(x10 ⁻⁵ m ² s ⁻¹)
Ammonia ^a	5.9x10 ¹	4100	1.5 (20 °C)	2.8
Carbon dioxide ^a	3.4x10 ⁻²	2400	1.91	1.4
Hydrogen sulphide ^b	1.0x10 ⁻¹	2100	1.36	1.5
Methane ^b	1.4x10 ⁻³	1700	1.84	1.6

^aSillen and Martell, 1964; ^bWilhelm et al., 1977; ^cPartitioning coefficient adapted from Sander (1999).

^d25°C unless otherwise stated. ^e27°C unless otherwise stated.

Table 3. Experimental studies on dissolved gas separation from natural and engineered anaerobic systems

Process/ Feed source	Target solute	Vol. (l)	SSA (m ⁻¹) /Length (m)	Contact time (min)	Hydraulic loading (m ³ m ⁻² min ⁻¹)	Temp (°C)	Flow rate (l h ⁻¹)		G/L ^a (-)	Concn. (mg l ⁻¹)		Reml. (%)	Ref.
							(Q _g)	(Q _l)		In	Out		
Spray aerator/ UASB effluent	CH ₄ S ²⁻	7.85	N/a /1.0 ^b	N/a	0.132	22	48.0-99.5	62.1	0.77-1.59	18 6	6 <1	73 97	Glória et al., 2016
Spray aerator/ Groundwater	CH ₄	7.39	N/a /0.95	N/a	N/a	N/a	N/a	N/a	5-30	N/a	N/a	45-55	Scherer et al., 2000
Packed column/ Groundwater	CH ₄	1238	175/ 4.5	N/a	0.22	10-11.5	550000	3600	153	1.5	0.006	99.6	Wessels, 2014
Tray aerator ^k / Groundwater	CH ₄	131 ⁱ / 219 ^f	6.7 ^k / 1.62 ^g	1.5-2.6	0.058	10-11.5	239000	3000	80	1.54	0.017	98.9	Wessels, 2014
Tray aerator/ Groundwater	S ²⁻	484 ⁱ / 730 ^f	119/ N/a	0.15	1.75	27.1-28	N/a	3160000	N/a	1.88	0.93	49.5	Duranceau et al., 2012
Membrane/ Groundwater	CH ₄	0.4 ^j	2545/ 0.2	0.08	2.5	10-11.5	1700	300	5.6	1.35	0.39	71	Wessels, 2014
Membrane ^l / UASB effluent	CH ₄	0.05 ⁿ	4600/ 0.1397	0.39	0.36	25	600	6	100	21	0.5	97.6	Cookney et al., 2016
Membrane ^m / AnMBR effluent	CH ₄	0.26 ^j	364/ 0.62	42	0.015	25	12	0.36	33	21	1.6	92.6	Cookney et al., 2016
Membrane ^l / UASB effluent ^e	CH ₄	0.4	1782/ 0.25 ^d	823	3x10 ^{-4,f}	37	Vacuum	0.029	94 kPa	18.9	1.9	89.9	Luo et al., 2014
Membrane ^l / UASB effluent ^e	CH ₄	0.07	2625/ 0.1397	1.2	0.12	25	79	0.63	125	30	0.45	98.5	Henares et al., 2016

SSA – Specific surface area (m² m⁻³). N/a – not available. UASB – Upflow anaerobic sludge blanket reactor. AnMBR – Anaerobic Membrane Bioreactor. ^aGas to liquid ratio.

^bStripping ratio (Eq. x). ^cDrop height. ^dMembrane fibre length. ^eSynthetic feed. ^festimate based on available dimensional information. ^gTotal flow length. ^hBased on maximum achievable G/L. ⁱFlooded tray volume. ^jShell-side priming volume. ^kLow profile aerator: specific surface area also generated through foam. ^lPolypropylene membrane.

^mPolydimethylsiloxane membrane. ⁿLumen-side priming volume.

Table 4. Advantages and disadvantages of desorption technologies for application to engineered anaerobic systems.

	Submerged aerators	Spray aeration	Packed column	Tray aerator	Membrane contactor
Surface area	Air bubble provides interfacial area	Water droplets form surface area	Packing media provide surface area ($250 \text{ m}^2 \text{ m}^{-3}$)	Trays provides basic interfacial area ($7 \text{ m}^2 \text{ m}^{-3}$)	Membrane provides surface area ($5000 \text{ m}^2 \text{ m}^{-3}$)
Hydraulic loading ($\text{m}^3 \text{ m}^{-2} \text{ h}^{-1}$)		2-7 (spray tower)	36-108	72 (low-profile)	1200
Description	<ul style="list-style-type: none"> Fine to coarse bubble diffusers immersed below depth >3 m Contactors placed in cascade (3-4 stages) Can use orifice plate to break down bubbles 	<ul style="list-style-type: none"> Droplets contacted with air Droplet size, velocity and drop height govern mass transfer Air turn-over needed 	<ul style="list-style-type: none"> High specific surface area media packed in column Water trickles downward on media Air passed upwards through media 	<ul style="list-style-type: none"> Gravity flow between sequential trays Thin film on tray provides contact Mixing encouraged in some designs in and between trays 	<ul style="list-style-type: none"> Hydrophobic material mediates non-disperse contact between aid and liquid Hollow fibre membranes packed in large numbers
Anaerobic application	<ul style="list-style-type: none"> CH_4, landfill leachate 	<ul style="list-style-type: none"> CH_4, UASB effluent 	<ul style="list-style-type: none"> CH_4, s/saturated g/water NH_3, return liquor 	<ul style="list-style-type: none"> H_2S, g/water 	<ul style="list-style-type: none"> CH_4, UASB/MBR effluent
Scale trialled	<ul style="list-style-type: none"> Full-scale for CH_4 	<ul style="list-style-type: none"> Full scale for THM^b Pilot scale for CH_4 	<ul style="list-style-type: none"> Full scale for THM^b Full-scale for CH_4 & NH_3 	<ul style="list-style-type: none"> Full scale for CH_4 (oil/gas) Full scale for H_2S (g/water) 	<ul style="list-style-type: none"> Full-scale for O_2, CO_2 Pilot scale for CH_4
Advantages	<ul style="list-style-type: none"> Simple Low fouling potential Can enable turn-up/turn-down Can achieve discharge consent^c Stripped air below LEL for methane^d 	<ul style="list-style-type: none"> Low ΔP_{air} permits use of low energy fans Simple installation 	<ul style="list-style-type: none"> Low ΔP_{liquid} Low ΔP_{air} is possible Can operate under turn-up/turn-down Good mass transfer High removal efficiency Methane recovery indicated with vacuum 	<ul style="list-style-type: none"> Smaller than packed tower – less hydraulic impact Easier accessibility for maintenance 	<ul style="list-style-type: none"> High mass transfer Constant mass transfer area simplifies design Sustain head pressure Very short HRT^e, small footprint Methane recovery demonstrated
Disadvantages	<ul style="list-style-type: none"> Variable surface area Poor mass transfer (long HRT^e and high energy) Methane re-use unlikely High aeration rates increase fouling^f Air compressor needed for air delivery 	<ul style="list-style-type: none"> Clogging of fine holes (can employ alternate geometry) Low/modest mass transfer Potentially high ΔP_{liquid} will increase energy 	<ul style="list-style-type: none"> Fouling results in inefficiency (regrowth) Clogging packing material from solids Flooding and foaming are considerable risks Channelling is possible 	<ul style="list-style-type: none"> Higher air flow requirements than packed column Limited hydraulic loading 	<ul style="list-style-type: none"> Requires solids pre-filtration ($\sim 10 \mu\text{m}$) to avoid clogging Without pre-filtration, fibre packing must be limited for clogging Membrane additional mass transfer resistance

^aFull-scale; ^bPilot-scale. ^cDischarge consent to closed conduit, generally 0.14 mg l^{-1} . ^dLEL – Lower explosive limit (5% v/v). ^eHRT – hydraulic residence time. ^fInorganic fouling.

^gTHM – Trihalomethane.

Table 5. Outlet gas phase composition from desorption technologies trialled for engineered anaerobic systems

	Scale	Packing	Driving force	Temp.	Contact time	G/L _{ratio}	S _{factor}	CH ₄ (diss.) removal	Gas composition					Ref.
									CH ₄	CO ₂	N ₂	O ₂	H ₂ S	
	(m ³)			(°C)	(s)	(-)		(%)	(%)				(ppmv)	
Packed Column/ UASB effluent	0.08 (0.035 ^a)	G2-sponge sheet	Air	10.5-28	7200	0.064 – 0.043	1.2- 1.8	70 ^c	39.4	5.0	55.5	<0.1	-	Matsuura et al., 2015
Diffused aeration/ Landfill Leachate	N/a	N/a	Air	N/a	2700 ^d	10-15	280- 420	99	0.028	-	-	-	-	Robinson & Carville, 2010
Spray aerator/ UASB effluent	0.004- 0.008	N/a	Air	22	0.5-1.0 ^e	0.77- 1.59	21.5- 44.5	73	2.5-5	-	-	-	100- 500	Glória et al., 2016
Membrane/ UASB effluent	0.0004	PDMS	Air/ Vacuum	16	19-377	0.77- 19.2	21.5- 545	72	0.028 ^f	-	-	-	-	Cookney et al., 2012
Membrane/ UASB effluent	0.0011	PE/PU	Vacuum	15-35	10080 - 33120	50-80 kPa	N/a	68-77	20-22	10-27	-	-	-	Bandara et al., 2011
Membrane/ Syn. effluent	0.0002	PP	Nitrogen	25	3-300	0.13- 100	3.6- 2800	55-97	0.2-20	-	-	-	-	McLeod et al., 2016

PDMS – Polydimethylsiloxane (nonporous membrane). PP – polypropylene (microporous membrane). PE/PU – Composite membrane comprised of non-porous polyethylene on porous polyurethane support. ^aSponge volume (44% of reactor void space). ^bFlowrate calculated based on internal volume. ^cBased on average. ^dEstimated from available data. ^eDrop height (m) which controls contact time. ^fBased on mass balance.

Table 6. Summary of destructive waste gas treatment technologies (Browell et al., 2009; GAL, 2009; Kent, 2012; Brandt et al., 2016).

	Description	Advantages	Disadvantages
Thermal oxidation	Ignition temperature for CH ₄ in air at 1 ATM, 625-672°C. Combustion efficiency dependant on temperature, pressure, volumetric composition of the fuel gas, size and shape of combustion chamber and the direction of flame propagation.	<ul style="list-style-type: none"> • Proven technology • Fast turn-up/turn-down for changes in flow (Turn-down 5:1) • Turn-up/turn-down through air-flow to respond to changes in composition (20-60% CH₄ v/v) • Low CV flare can treat 10-50% CH₄ v/v 	<ul style="list-style-type: none"> • Waste gas mixture must be 5-15% for ignition • Need adequate oxygen to complete combustion • Blending air to achieve 5-15% introduces Health and safety risk • Need sufficient heat/ energy to sustain combustion • By-products formation (CO, NO_x) • Considerable capital cost
Non-catalytic thermal oxidation (Regenerative thermal oxidation)	Methane oxidised to CO ₂ on a hot reaction matrix (e.g. ceramic). Turbulence, residence time and temperature govern efficiency.	<ul style="list-style-type: none"> • Operates auto-thermally from 0.3-0.8% v/v CH₄ (auto-oxidation temperature CH₄, 540°C) • >0.8-27% v/v CH₄ requires dilution with air • High destruction efficiency 	<ul style="list-style-type: none"> • Mature for industrial air treatment of Volatile Organic Compounds (VOCs) but not widely applied for anaerobic waste gas • High capital cost and installation cost
Biofiltration	Specialist methane oxidising micro-organisms colonise packed bed media	<ul style="list-style-type: none"> • Good methane oxidation from 0.17 to 20% CH₄ v/v off-gas demonstrated • Potential technology for large gas flows • Combined H₂S and CH₄ removal has been proposed • Lower capital cost versus RTO and flare • Lower operational costs versus RTO and flare 	<ul style="list-style-type: none"> • Requires pH, oxygen, nutrient control • Moisture level must be maintained in packed bed • For large flows, scale restrictions may apply (like odour treatment) • Limited experience at full-scale; further development needed

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Heile, S.

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