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Review

Gas transport in landfill cover system: A critical appraisal

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ABSTRACT

Landfill gas (LFG) emission is gaining more attention from the scientific fraternity and policymakers recently due to its threat to the atmosphere and human health of the populace living in surrounding premises. Though landfill cover (LFC) (viz., daily, intermittent and final cover) is widely used by landfill operators to mitigate or reduce these emissions, their overall performance is still under question. A critical analysis of available literature, primarily pertaining to (i) the composition of the landfill gases and their migration in the LFC system, (ii) experimental and mathematical investigations of the transport mechanism of gas and (iii) the impact of additives to cover soils on transport and fate of gas, has been conducted and presented in this manuscript. Investigation of the efficiency of modified soil was mainly focused on laboratory test. More field tests and application of amended cover soils should be conducted and promoted further. Studies on nitrous oxide and emerging pollutants, including poly-fluoroalkyl substances transport in landfill cover system are limited and need further research. The transport mechanisms of these unconventional contaminants should be considered regarding the selection of LFC materials including geomembrane and geosynthetic clay liners. The existing analytical and numerical models can provide a basic understanding of LFG transport mechanisms and are able to predict the migration behaviour of LFG; however, there are still knowledge gaps concerning the interaction between different species of the gas molecule when modeling multi-component gas transport. Gas transport through fractured cover should also be considered when evaluating LFG emission in the future. Simplified design method for landfill cover system regarding LFG emission based on analytical models should be proposed. Overall, mathematical models combined with experiments can facilitate more visualized and intensive insights, which would be instrumental in devising climate adaptive landfill covers.

1. Introduction

Greenhouse gas emission was one of the prime agendas of discussion in the recently conducted COP26 at Glasgow (Scotland), where world leaders concentrated on mitigating global warming. In this context, landfills are one of the largest sources of greenhouse gas, which is responsible for 11% of global anthropogenic methane (CH_4) (Jung et al., 2019) and several other landfill gases (viz., CO_2, N_2O, NH_3) and non-methane volatile organic compounds (NMVOCs) that are generated during the decomposition of organic matter of municipal solid wastes (MSW). It is indicated that emission flux of methane ($\sim 4.50 \times 10^1$ to 4.15×10^4 g/m^2/day) was the highest, followed by nitrous oxide ($\sim 2.50 \times 10^{-3}$ to 3.75×10^1 g/m^2/day), and NMVOCs ($\sim 2.00 \times 10^{-3}$ to 7.32×10^{-1} g/m^2/day) in landfill (Manheim et al., 2021). Unfortunately, despite having regulations to reduce the landfilling of MSW, a significant amount of this waste is managed by this method worldwide. For instance, in the European Union (European Commission, 2006; European Environmental Agency, 2013; Eurostat, 2015), Russia (Starostina et al., 2014), USA (Tonjes and Greene, 2012; Paleologos et al., 2016) and India (Singh et al., 2011; Ramachandra et al., 2018; Pujara

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et al., 2019), the percentage of landfilled MSW is 64%, 90%, 60% and 90%, respectively. Also, generation rate of MSW is approximately increasing at 40% and 19% for developing and developed countries (Kaza et al., 2018), respectively, resulting in generation of 3.40 billion tonnes of waste by 2050.

Landfill cover (LFC), including interim or final covers, passively or actively ventilated biofilters, biowindows and daily used biotraps are most promising and cost-effective options when other treatments (i.e., combined heat and power plant and controlled flaring) are not feasible due to low concentration of methane and gas flow rate (Huber-Humer et al., 2008). LFC system can be classified into (i) daily- (layered after the landfilling of the working face every day) (Spokas and Bogner, 2011), (ii) intermediate- (a temporary cover for a waste area without usually landfilling for more than one year) (Spokas and Bogner, 2011) and (iii) final cover (constructed on the waste when the landfilled waste reaches the designed height of the landfill). Recently, in the aerobic zone of LFC, methanotrophic microorganisms, such as type I (mainly Methylocaldum) and type II (mainly Methylocystis) methanotrophs, accelerates the oxidation of methane to carbon dioxide, which has less global warming potential (Moon et al., 2010). Furthermore, studies have been performed to augment the mitigation properties of LFC by directly or indirectly accelerating the oxidation capacity by adding additives, such as earthworm cast, mature compost, woodchips, biochars, activated carbon, pine bark, shredded rubber, slags from steel industries, etc. (Moon et al., 2010; Niemczyk et al., 2022). These additives are instrumental in (i) providing higher surface area and nutrients for the proliferation of methanotrophs, (ii) enhancing the air permeability, and (iii) maintaining the proper moisture content and pH of the LFC system.

It should be recalled here that LFC represents a biogeochemically active multiphase porous media with its primary constituents (Aghdam et al., 2019; Duan et al., 2021) (i) degradable and non-degradable solid fractions, which contain pores, and (ii) these pore spaces are filled with multicomponent landfill gases (viz., CH_4, CO_2, NH_3 N_2O, H_2S and NMVOCs), moisture and microorganisms. Hence, the migration of LFG in LFC represents a reactive transport phenomenon in porous media, which has to be studied thoroughly by considering the principals and theories of multiphysics coupling. A few encouraging laboratory experiments, field tests, mathematical simulations have been performed by previous researchers to understand the mechanism of LFG transport in LFC systems. Also, there are bunch of existing reviews concerning the origin and nature of crude oil volatile emissions (Rajabi et al., 2020), the treatment methods of landfill contamination (Omar and Rohani, 2015), the ways of MSW treatment (Anshassi et al., 2021), the different technical quantification and control options of LFG (Huang et al., 2022), and the coupled behaviour of wastes in landfills (Lu and Feng, 2020). However, the numbers of these studies are very limited to conclude the overall behaviour in terms of physico-bio-chemico-thermo-geomechanical properties of LFC both for short- and long-term basis. The mechanism of LFG transport inside landfills and cover system hasn't been included in the above-mentioned reviews, which is crucial for the understanding of gas emissions.

It is the need of the hour to understand the reactive transport phenomenon in a biogeochemically active multiphase porous media to develop strategies to mitigate or reduce the gaseous emission from the landfill to protect the environment and human health living in surrounding premises. This necessitates a critical analysis of the available literature to understand the (i) transport phenomena of gases in LFC and parameters that influence it, (ii) impact of additives on the variation of overall properties of LFC including methanotrophic communities and (iii) long term performance of LFC. It is believed that this exercise would be instrumental in devising a climate-adaptive cover system, which is the utmost requirement for landfill operators.

2. Landfill gas transport in cover system

2.1. Compositions of landfill gas

LFG is mainly composed of 40–60% of methane, approximately 40% of carbon dioxide, nitrous oxide (N_2O), and more than 100 types of non-methane volatile organic compounds (NMVOCs) (Osra et al., 2021; Duan et al., 2021), which are generated during the microbially induced biochemical decomposition of organic matter in a landfill, as depicted in Fig. 1. In general, the generation of LFG continues for several decades, except for bioreactor landfills, where decomposition is accelerated to reduce the treatment time (Chembukavu et al., 2019; Mohammad et al., 2021a, b, 2022), until the majority of organic compounds in the wastes were decomposed completely. The generated LFG can be collected by the gas recovery system, oxidized by the microorganism, and adsorbed by the porous medium in the landfill (Sun et al., 2015). The rest of the quantity of LFG will be emitted into the atmosphere, which is responsible for the greenhouse effect due to presence of methane, carbon dioxide, and nitrous oxide in it. The concentrations and emission rates of CH_4 and N_2O from some landfills in different countries are listed in Table 1. It is indicated that the emission rate of N_2O is 1–3 orders of magnitude less than the methane. The global warming potentials of methane and nitrous oxide are 28–36 and 265–298 times that of carbon dioxide for a 100-year timescale (US EPA, 2020). The most noteworthy is that the emission of N_2O, generated from nitrification, incomplete denitrification and chemo-denitrification (Ishigaki et al., 2016), was usually neglected by many researchers while investigating the gas emission from the landfill because landfills are widely known as an emission source of methane. Besides, the Intergovernmental Panel on Climate Change (IPCC) does not require waste landfills to report the emission of nitrous oxide in national inventories. Simultaneously, the emission of N_2O is highly dependent on environmental conditions such as the concentrations of oxygen and nitrite (Kampschreur et al., 2008). Therefore, the transport of N_2O in the LFCs has hardly been investigated though it is one of the most important greenhouse gases.

A field test conducted by Harborth et al. (2013) indicated that the concentration of N_2O tested in the hotspots of a landfill working face in Germany can reach up to 24,000 ppm, which implies that its emission cannot be overlooked. Ishigaki et al. (2016) conducted a field test in landfills and found that the percentage of N_2O and CH_4 in LFG are 6.7% and 31%, respectively. In addition, ammonia, hydrogen sulfide, and other traces of VOCs such as methyl mercaptan, ethanethiol emitted into the atmosphere will create severe odour nuisance and acute irritation of human organs (Wu et al., 2018; Wang et al., 2019; Mohammad et al., 2021c).

It is indicated that concentrations of methane from hotspots can be several orders of magnitude greater than the limit standard. Therefore, controlling methane emission from landfills is still a difficult task, if not impossible, in both developed and developing countries. The concentration and types of NMVOCs in some landfills from all around the world are presented in Table 2 and Fig. 2 (refer to Table 2 for the abbreviation of landfill name). It is indicated that VOCs concentration at different landfills differed greatly. For instance, the total VOC concentration at Izmir landfill (IZM) in Turkey can be four orders of magnitude less than that at Sudokwon landfill (SDK) in South Korea, which indicates that VOC emission from landfill is strongly related to the variation of climate, geographical regions and waste characteristics. It can also be deciphered from Table 2 that the concentration of hydrogen sulfide emitted from landfills can be 2–5 orders of magnitude greater than the odour threshold (≈0.57 μg m^{-3}) mentioned by Nagata and Takeuchi (2003), which can lead to severe odour nuisance to the surrounding residents.

In terms of greenhouse gases, the proportion of methane in the global warming effect of landfills by CH_4 and CO_2 emissions is about 96–98% and 2–4%, respectively (Molino et al., 2013). The effect of VOCs on the human health and environment is pernicious (can be much more severe than CH_4 and CO_2), although their proportion only accounts for ≤1–2% of landfill gas (ATSDR, 2007; Chiriac et al., 2007; McKenzie et al., 2012).

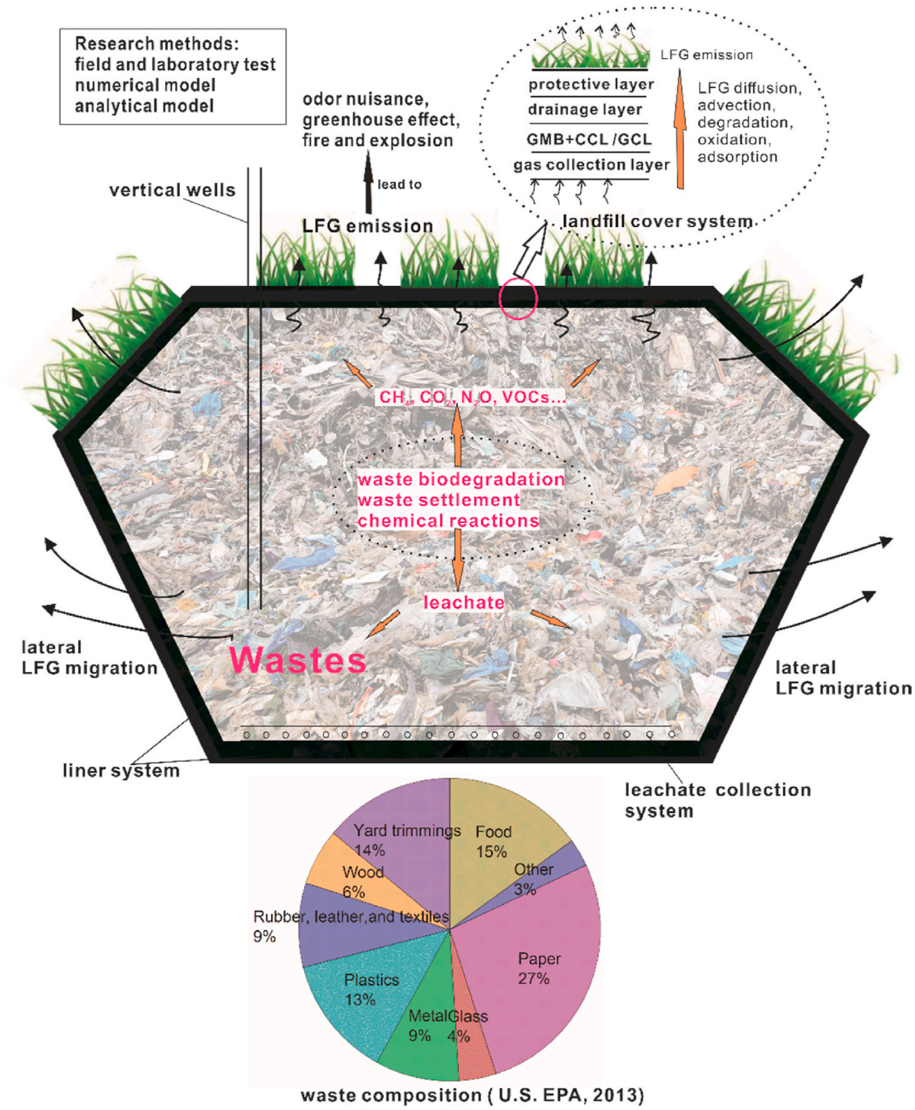


Fig. 1. Conceptual scheme of landfill gas generation and emission.

Continuous exposure to the LFG consisting of chlorinated hydrocarbons and aromatic hydrocarbon can lead to serious health issues for human beings, particularly on landfill workers (Zou et al., 2003), which can not only cause acute irritation of human organs, headache, and difficulty of focusing (Blount et al., 2006), but also increase the risk of cancer (European Environment Agency, 2008). Therefore, development of mitigation strategy for methane and VOCs emitted from landfills is a worldwide concern (Huang et al., 2020; Randazzo et al., 2020).

Recently, emerging LFG emission, including some per- and polyfluoroalkyl substances (PFAS) has gained much attention. PFAS are persistent and bioaccumulate in the environment, and their concentration increases with time in the organs and blood, leading to a great threat to human health, even at low concentrations (Mukerji et al., 2015; Kong et al., 2019). Ahrens et al. (2011) investigated polyfluoroalkyl compounds (PFCs) emission to air from wastewater treatment plants and two landfills in Ontario, Canada. It is indicated that in Ontario, fluorotelomer alcohol (FTOH) accounts for 93–98% of the total PFCs, and the concentration of FTOHs was 2780–26,430 pg/m^3. The concentration of 8:2 FTOH at landfill site ranges from 1290 to 17,380 pg/m^3. FTOHs were also detected in a closed and active sanitary landfill in Northern Germany (Weinberg et al., 2011). However, studies of PFAS emission from landfills are limited and more investigations should be conducted on the emission and fate of emerging pollutants, including PFAS from landfills.

2.2. Landfill cover system

Initially, the LFC system usually consisted of thick local soil layer placed over the MSW and was built to minimize rainfall infiltration and LFG emission. In the later stage, the hydraulic barrier gradually developed into a geosynthetic clay liner (GCL) or a compacted clay liner (CCL) overlaid by a geomembrane in laboratory and field studies. GCL usually consists of a thin layer (total thickness ≈5–10 mm) of bentonite or other low permeability material sandwiched between geotextiles and/or geomembranes, mechanically held together by needling, stitching, or chemical adhesives (Bouazza, 2002; Rouf et al., 2016; Bouazza et al., 2017; Rowe, 2020; Wang et al., 2021) and convenient and affordable to install (Bouazza, 2002). Geomembranes, which are made of linear low-density polyethylene (LLDPE), high density polyethylene (HDPE), polyvinyl chloride (PVC) and very low-density polyethylene (VLDPE), are relatively thin (normally ≤2.5 mm) and commonly used together with compacted clay and/or a GCL. For landfill applications, the primary function of a GMB is to provide a diffusive barrier to inorganic contaminants and prevent the advective flow of contaminants through the liner. Generally, constructing with geosynthetics in internal drainage layers is also included in modern landfill covers. In the 90s of

Table 1
Methane and nitrous oxide emission from landfills.

Gas	Landfill location	Measuring location	Concentration[a] (ppm)	Flux[b] (mg/m^2/min)	Reference
Methane	Xi'an, China	Final cover	2–2143	NM	Shen et al. (2018)
		Temporary cover hotspots	15.42–33975.24		
	Zealand, Denmark	Final cover hotspots	>3000	NM	Scheutz et al. (2011)
	Delhi, India	Soil cover surface	NM	9.2–60	Chakraborty et al. (2011)
	Guwahati, India	NM	NM	28.7–143.1	Gollapalli and Kota (2018)
	Palermo, Italy	Edge of the landfill top surface	NM	0.06–776.94	Di Trapani et al. (2013)
		HDPE temporary cover	NM	0.06–390	
	Bellolampo, Italy	The whole landfill site	NM	0.06–483	Di Trapani et al. (2013)
	Makassar, Indonesia	An open dump disposal site	32.4–1069.9	5.8–456.7	Lando et al. (2017)
	Saitama Prefecture, Japan	Semi-aerobic landfill surface	1.5–5	2×10^{-4}	Izumoto et al. (2018)
	South of Germany	Final cover with a 90-cm compost layer	2.8–6	NM	Zhu et al. (2013)
	Ho Chi Minh City, Vietnam	Landfill ambient air	0–506.8	NM	Bui and Nguyen (2020)
	Falköping, Sweden	The whole landfill site	NM	2.52	Galle et al. (2001)
	Nashua, U.S.	The whole landfill site	NM	54.15	Mosher et al. (1999)
	Palermo, Italy	Landfill surface	NM	24.6	Di Bella et al. (2011)
	Chanthaburi, Thailand	Waste piles		5.09–18.67	Wangyao et al. (2021)
Nitrous oxide	Southeast of Sweden	sewage sludge cover		0–0.017	Börjesson and Svensson (1997)
		Mineral soil cover		−0.0001 -0.595	
	Helsinki, Finland	Organic soil cover with gas collection system		0.1	Rinne et al. (2005)
	Pohlsche Heide, Germany	Working face covered with a compacted layer (10–15 cm) of bottom ashes from inciner-ation of refuse derived fuel.	24,000 in hotspots	max. 7.13	Harborth et al. (2013)
	Ningbo, China	Landfill soil covers		0.057	Long et al. (2018)
	Chanthaburi, Thailand	Waste piles		0.11–1.86	Wangyao et al. (2021)

[a] According to the Carbon Farming Initiative (2013), the permitted CH_4 concentration limit for final landfill should be less than 500 ppm.
[b] According to the Carbon Farming Initiative (2013), the allowable CH_4 emission rate through a final landfill cover should be less than 41.67 mg/m^2/min.

the last centuries, the alternative earthen final cover (AEFCs) was developed, which is similar to the original soil covers, and the water storage capacity of finer-textured soils was exploited by the thick layer of fine grain soil, and is considered worldwide (Zhan et al., 2016, 2020; Ng et al., 2016; Zhang et al., 2020; Chetri and Reddy, 2021). The capacity of methane removal efficiency in traditional landfill cover soil was limited by many factors such as water retention capacity, organic content, and methanotrophic bacteria, etc. (He et al., 2012; Sadasivam and Reddy, 2014). Therefore, the potential for increasing methane oxidation by using alternative landfill covers has been widely investigated by researchers around the world (He et al., 2012; Yargicoglu and Reddy, 2018). Attention has been paid to enhance methane oxidation and sorption capacities by using organic materials (e.g., sewage sludge, composts, biochar) either as amendments or alone to the cover soil.

Typically, a surface layer, a soil protective layer, a drainage layer, a hydraulic barrier, a gas collection layer, and a foundation layer from the top down were included in modern landfill cover systems according to US EPA (US EPA, 2004). Materials used for landfill cover system construction include sand, gravel, CCLs, geomembranes (GMBs), GCLs, etc. These barrier materials can be used alone (such as GMBs, CCLs, and GCLs) or in combination as a composite cover. It has been shown that a composite barrier involving GMB/GCL, GMB/CCL, or GMB/GCL/CCL in the cover system allows less infiltration and LFG emission than a LFC with a single GMB, GCL, or CCL (Bonaparte et al., 2004; Xie et al., 2016, 2017, 2018). However, the later cover system is more affordable than the former.

2.3. Mechanism of landfill gas transport in cover system

In order to get good control of LFG emission, understanding the mechanism of its transport through landfill cover system material is of great importance (Joseph et al., 2019). The migration of landfill gas in LFC systems is affected by several factors, including the (i) type of cover system and its characteristics (i.e., moisture content, density, saturation, intrinsic permeability, etc.), (ii) partial pressure, concentration and water dissolution coefficient of LFG, and (iii) climatic conditions (temperature, atmospheric pressure, infiltration) (Rowe et al., 2004; Xie et al., 2016; Rouf et al., 2016; Majdinasab et al., 2017). Depending on the mentioned factors, the migration mechanism of LFG in LFC can be either advection, diffusion, sorption, degradation, and oxidation or a combination of them, as illustrated in the following.

2.3.1. Diffusion and advection of LFG in cover system

Diffusion is a physical process of mass flow and molecular exchange where gas movement occurs due to Brownian motion and concentration gradient through the air-filled pores in unsaturated porous media (Troeh et al., 1982; Aubertin et al., 2000; Barral et al., 2010). Due to differential gas concentration between landfill system and atmosphere, LFG tends to diffuse through the LFC system from the high concentration zones to the low concentration ones. The air-filled pores in porous media are maximum at fully dry and minimum at fully saturated conditions. When the porous medium is highly saturated (>85%), the LFG migrates partly through the gas phase, and it also migrates in the liquid phase by dissolving in the pore water (Aubertin et al., 2000; Bouazza and Rahman, 2007).

Fick's laws can be used to describe mass diffusive flux of LFG through LFC systems (Aubertin et al., 2000; Bouazza and Rahman, 2007; Rouf et al., 2016) as presented in Eq. (1) for the one-dimensional diffusion:

$$J = -D_e \frac{\partial C_g}{\partial z} \tag{1}$$

where z is a distance (m), D_e is the effective diffusion coefficient of LFG (m^2·s^{-1}), J is the diffusive flux of LFG (g·m^{-2} s^{-1}), and C_g is the concentration (g·m^{-3}).

The transient one-dimensional gas diffusion equation (also called

Table 2
Composition and concentration in a few typical landfills around the world (μg/m^3).

	Benzene	Toluene	Ethylbenzene	Hydrogen sulfide	Carbon disulfide	Ammonia	Chlorobenzene	Limonene	Ethanethiol	Methanthiol	Heptanal	Dimethyl sulfide	Hexanal
A landfil in Beijing (BL)[a]	12	33	55	46.3	8.3	612.5	53.8	63.5			14.3	9.1	13.3
Asuwei landfill in Beijing (ASU)[b]	3.1–62	5.3–166	3.8–134	31–867	1.9–31			36–1214		16–149		8.9–102	
Tianziling landfill in Hangzhou (TZL)[c]	3.82	60.4	23.3	514.52	0.66	3960	1.28		0.48	5.3	0.12	18.52	
Guangzhou Datianshan (DTS)[d]	1.2–167	1.7–202	0.1–52				0.1–3.7	0.1–162			0.1–3.1		0.3–2.8
A landfill in France (FL)[i]		2190	40					1470–1870					
Mallorca Son Reusin Spain (MSR)[f]	7.8–15.9	58.8–80.8	1.76–3.31	800		300	0.35–0.89	N.D.					6.15–7.88
Deonar landfill in India (DEO)[j]	286.1	70.5	0.5										
Bologna landfill in Italy (BOL)[h]		0.79–1.62	0.23–0.5				0.07–0.14	0–13.44			0.01–0.02		0.01–0.12
Izmir andfill in Turkey (IZM)[g]	0.29–0.53	4.8–18.9	0.45–3.03				0.0009–0.12				0.16–0.64		0.59–1.01
Sudokwon in South Korea (SDK)[e]	21,400	82,150	19,540	21,050						23,800			
Landfill in Japan (JL)[k]	21,900	16,700	18,700					80					
Landfill in Poland (PL)[l]	270	2970	680					2360					
Odense Landfill in Denmark (New) A (ODN)[m]				50,600-276,0000									
Odense Landfill in Denmark (Old) (ODO)[m]	5100–161,000	13,400-161,0000	7300–73,200										

[a] Wu et al. (2017).
[b] Duan et al. (2014).
[c] Ying et al. (2012).
[d] Zou et al. (2003).
[e] Shin et al. (2002).
[f] González et al. (2013).
[g] Dincer et al. (2006).
[h] Davoli et al. (2003).
[i] Chiriac et al. (2007).
[j] Majumdar and Srivastava (2012).
[k] Takuwa et al. (2009).
[l] Sadowska-Rociek et al. (2009).
[m] Duan et al. (2021).

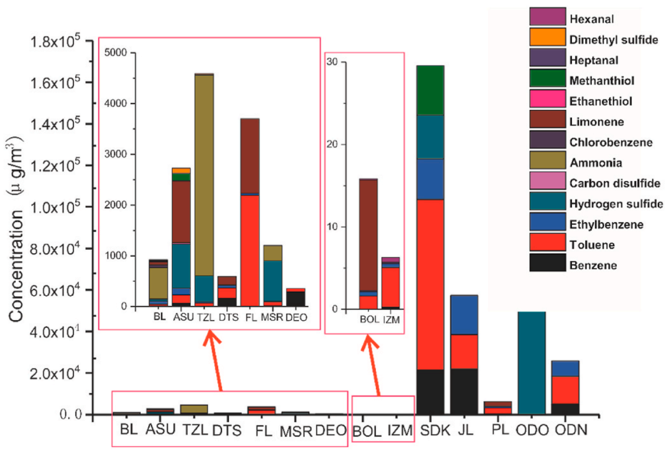


Fig. 2. VOC concentration and composition in different landfills around the world.

Fick's second law) can be obtained by Fick's first law and continuity equation as presented in Eq. (2) (Rowe et al., 2004):

$$\theta_a\left(\frac{\partial C_g}{\partial t}\right) = D_e\left(\frac{\partial^2 C_g}{\partial z^2}\right) \tag{2}$$

where θ_a is the air-filled porosity in the porous medium (m^3/m^3); t is the time (s). Eq. (2) can also be used to calculate the diffusion coefficient of the gas in porous media, which depends on the pores and fluid properties, such as degree of saturation, total porosity, tortuosity, molecular mass and the diffusion coefficient of the gas in the atmosphere. The effective area available for flow in the cross-section of the porous medium, the bending factor, is the main determinant (Millington, 1959).

In advection, the LFG moves from a region with higher total pressure to one with lower total pressure until the pressure equilibrium is reached in the two regions (Troeh et al., 1982). Increased total pressure and corresponding partial pressure can be resulted due to the generation of LFG in landfills, which may lead to gas advection. The differential air pressure takes place in the landfill due to the natural fluctuation of the surface-atmosphere of the LFC system and the pressure difference caused by the gas accumulation at the bottom of the cover system (Vangpaisal and Bouazza, 2004). Groundwater table or temperature changes induced by leachate generation and microbial decomposition can also lead to pressure differential at the same time (Lundgren, 2001; Vangpaisal and Bouazza, 2004). Moreover, the orientation of the gas flow towards the forced-extraction wells can also be explained by pressure gradient and advection.

A number of studies have shown that the migration of advective gases through porous media with low permeability can be estimated by Darcy's law, as presented in Eq. (3) (Alzaydi et al., 1978; Vangpaisal and Bouazza, 2004). Meanwhile, Massmann (1989) showed that when the pressure difference was <50 kPa, the groundwater flow model (as shown in Eq. (3)) can be used to describe advective gas migration in the landfill soil covers.

$$Q = -\frac{k}{\mu}A\frac{dP}{dz} \tag{3}$$

where k is the intrinsic permeability of the porous media (m^2), Q is the gas flow through the cross-section of the porous medium ($m^3{\cdot}s^{-1}$), μ is the dynamic viscosity coefficient ($N{\cdot}s{\cdot}m^{-2}$), A is the cross-sectional area of the porous material (m^2), and dP/dz is the pressure gradient.

Gas permeability coefficient K ($m{\cdot}s^{-1}$) through porous media can be estimated by Eq. (4).

$$K = \frac{\rho g}{\mu}k \tag{4}$$

where ρ is the density of the gas ($kg{\cdot}m^{-3}$), and g is the gravitational acceleration ($m{\cdot}s^{-2}$).

Further considering the compressibility of gas, modified Darcy's law is used (Ng et al., 2015a, 2015b):

$$Q = \frac{kA\left(P_{in}^2 - P_{out}^2\right)}{2\mu H P_{out}} \tag{5}$$

where; H is the thickness of soil (m); P_{in} and P_{out} is absolute gas pressure (Pa) at inlet and outlet, respectively.

2.3.2. Sorption, degradation and oxidation of LFG

Sorption has a great influence on the reduction of LFG by retaining methane, NMVOCs, ammonia, and hydrogen sulfide in LFC systems. For instance, VOCs removal efficiency can be greatly influenced by the degradation and adsorption process in LFC systems (Rajamanickam et al., 2017; Raga et al., 2018). The water content of the soil is significant to the adsorption of VOCs because polar water molecules are strong competitors at the adsorption sites of soil particles relative to non-polar organic compounds (Rhue et al., 1989; Ong and Lion, 1991). In the case of high-water content, the adsorption process of VOCs in the soil can be mainly divided into five ways (Ho and Webb, 2006): (i) adsorbed by the gas-water interface; (ii) dissolved in pore water of soils; (iii) dissolved VOCs from pore water absorbed by soil particles; (iv) VOCs decompose from the liquid phase to organic matters and (v) VOCs condense into pores, as demonstrated in Fig. 3. The VOCs directly sorbed on the soil particles and decomposed into the organic matter may compete with the water molecules adsorbed on the soil particles when the moisture content of the soil is low. At this time, the adsorption of VOCs occurs on the surface of soil particles. The sorption capacity of soil to VOCs mainly depends on the surface area, organic matter content, and water content of soil (English and Loehr, 1991). Meanwhile, researchers have indicated that the adsorption capacity of soil with low water content depends on the surface area of soil (Chiou and Shoup, 1985; Rhue et al., 1988).

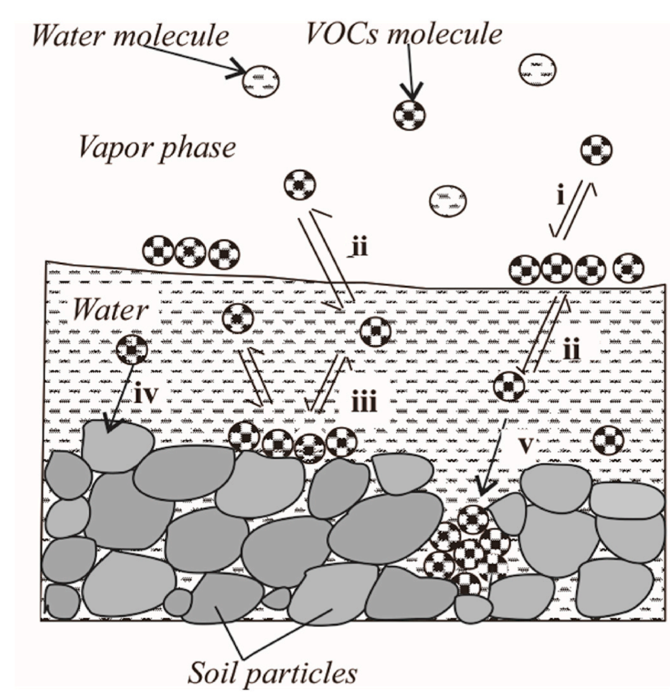


Fig. 3. VOCs sorption with a film water on the surface of soil; Mechanisms: i-sorption at the gas-water interface; ii-dissolution; ii-adsorption onto the mineral surface from the aqueous phase; iv-partitioning into the organic matter; v-VOCs condensation into pores.

Assuming that the conversion partition is linear superposition, the sorption retardation factor R can be expressed as Eq. (6) (Brusseau et al., 1997; Kim et al., 2005).

$$R = \beta_w + \beta_g + \beta_d + \beta_i \tag{6}$$

where β is the partial retardation factor with the subscripts w, g, d and i referring to VOCs retention in the aqueous phase, in the gaseous phase, sorbed at the solid domain of the soil and at the air-water interface, respectively, and they can be mathematically represented by Eqs. (7)–(11).

$$\beta_w = \frac{\theta_w}{K_H \theta_a} \tag{7}$$

$$\beta_g = 1 \tag{8}$$

$$\beta_d = \frac{\rho_b K_d}{K_H \theta_a} \tag{9}$$

$$\beta_i = \frac{a_i K_i}{\theta_a} \tag{10}$$

with:

$$K_i = \frac{\Gamma_i}{C_g}; \quad K_H = \frac{C_g}{C_w}; \quad K_d = \frac{C_s}{C_w} \tag{11}$$

where K_i is the interfacial adsorption coefficient ($cm^3 cm^{-2}$); K_H is Henry's (law) constant ($cm^3 cm^{-3}$); K_d is the sorption coefficient ($cm^3 g^{-1}$); C_s is the VOC concentration ($mol \bullet g^{-1}$) adsorbed by the solid phase; C_w is the VOC concentration ($mol cm^{-3}$) in the aqueous phase, and Γ_i is the VOC concentration ($mol \bullet cm^{-2}$) adsorbed at the air-water interface.

Landfill gas is decomposed by aerobic or anaerobic microorganisms in the cover system during migration. Soil hosts a large number of microorganisms, which is an excellent natural biological filter and a variety of different VOCs adsorbents for degradation (Insam and Seewald, 2010). The precondition of VOC degradation by microorganisms is that VOCs are dissolved in water-filled pores of soil or adsorbed by solid soil particles because VOCs cannot be in contact with microorganisms in a gaseous state (Rivett et al., 2011). The degradation rate of LFG in the landfill cover systems depends on many factors, including water content, temperature and distribution of nutrient content of soil, presence of pathogens, and microbial activity, etc. (Sadasivam and Reddy, 2014). An important landfill design method is to reduce the release of LFG by improving its degradation rate (Scheutz et al., 2009; Tassi et al., 2009).

When studying degradation of contaminants through porous media, the degradation of LFG is usually simulated by a first-order degradation model (Jury et al., 1990; Beyer et al., 2007):

$$\frac{\partial C_g}{\partial t} = -\lambda C_g \tag{12}$$

where λ is the first-order degradation rate of LFC (s^{-1}).

As the anaerobic- and aerobic bacteria exist in the soil, the final product of anaerobic metabolism will act as the nutrient to the aerobic metabolism and eventually decompose into water and CO_2 (Owen et al., 2007). Utilizing oxygen that diffuses from the atmosphere into the cover, methane can be oxidized by methanotrophic microorganisms in waste or soil materials in the landfill as described by the following reaction:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O + \text{heat} \tag{13}$$

The concentrations of both oxygen and methane have a significant impact on the activity of methanotrophic bacteria because most of them are strict aerobes and obligate methanotrophs (Hanson and Hanson, 1996). Therefore, methane oxidation in LFC system can be described by a general-purpose model, which includes a dual-substrate kinetic model as mentioned in Eq. (14) (Ishiwata, 1998; De Visscher and Van Cleemput, 2003):

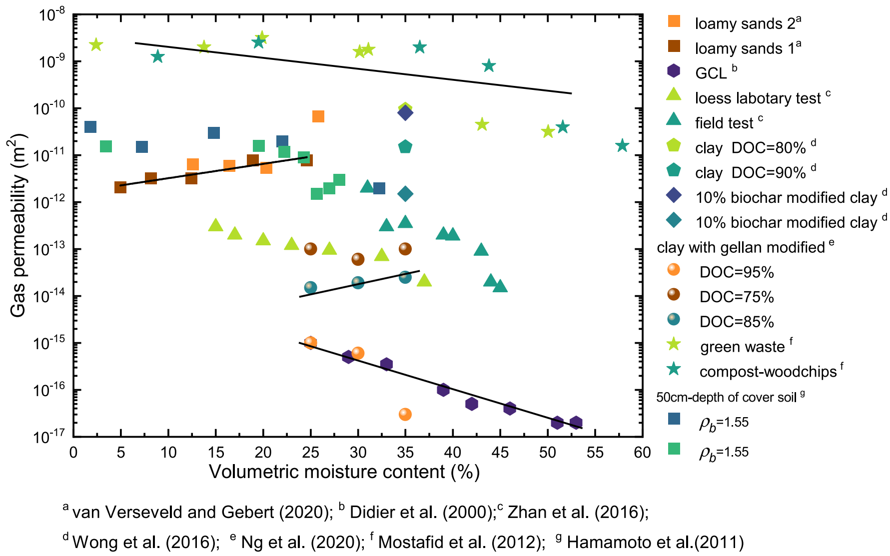
$$r_{CH_4} = -\frac{V_{\max} C_{CH_4}}{K_m + C_{CH_4}} \frac{C_{O_2}}{K_{O_2} + C_{O_2}} \tag{14}$$

where r_{CH_4} is the methane oxidation rate; K_m and K_{O_2} are the Michaelis-Menten constants for methane and oxygen, respectively; K_m and K_{O_2} are the concentration of methane and oxygen, respectively; and $V_{\max}$ is the maximum methane uptake rate.

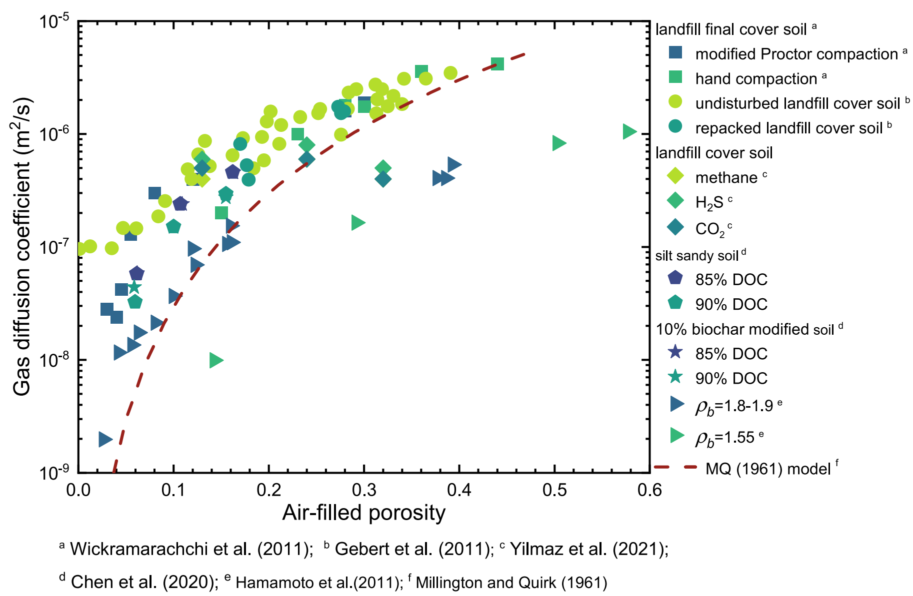
3. Experimental assessment

3.1. Gas advection and diffusion in cover material

Laboratory tests of gas diffusion and advection through cover materials, including soil, GCL, and GMB, have been conducted by many researchers (McWatters, 2015; Zhan et al., 2016; Rouf et al., 2016; Garg et al., 2019; Yilmaz et al., 2021). Advection of LFG through GMB involves its migration through tears, holes, or flaws in a geomembrane (Rowe et al., 2004). Intact geomembranes refer to the GMB without any defects or wrinkles, which can provide an excellent barrier to gas advection. However, LFG can still transport through GMB by diffusion process (Rowe et al., 2004; Bouazza, 2021). Furthermore, the soil has been modified by additives, including biochar (Wong et al., 2016; Garg et al., 2019; Chen et al., 2020; Chiu and Huang, 2020), compost (Mostafid et al., 2012), woodchips (Pokhrel et al., 2011; Mostafid et al., 2012), sludge (Xue et al., 2016; Qin et al., 2020), and lime (Yunmin et al., 2018) and used as an alternative landfill cover material. Biochar is an organic, highly porous material derived from plant, manure, or wood-based biomass by pyrolysis in a limited oxygen environment (Lehmann and Joseph, 2015; Sadasivam and Reddy, 2015a). Fig. 4 shows LFG permeability and diffusion coefficients through different types of soils obtained through laboratory and field test. It can be concluded that permeability and diffusion coefficient of LFG through cover material is strongly related to the type of material, the moisture content, soil compaction, air-filled porosity, vegetation coverage, and the modification of soil. Moisture content and degree of compaction can be the two guiding factors affecting LFG transport. It is indicated from Fig. 4a that gas permeability of cover material ranges from 10^{-17} to 10^{-8} m^2. Permeability of loamy sands ranges from 0.1×10^{-11} m^2 to 2.6×10^{-11} m^2 when dry density (ρ_b) of soil ranges from 1.3 g cm^{-3} to 1.8 g cm^{-3} (van Verseveld and Gebert, 2020). Gas permeability of 15% biochar modified soil decreased by two orders of magnitude when its compaction degree increased from 80% to 90% (Wong et al., 2016). Typically, gas permeability decreases with the increase of moisture content since the soil pores for air flow are reduced and occupied by the water. Fig. 4a shows that when the volumetric moisture content of loess increased from 15% to 35%, gas permeability decreased by one order of magnitude (Zhan et al., 2016). However, there are studies found that the increasing trends of permeability with the moisture content (Ng et al., 2020; van Verseveld and Gebert, 2020). It was explained by the formation of different soil fabrics in biopolymer modified soil, especially in the fine-grained soils. The pore size distribution at the same void ratio is different. Therefore, increasing gas permeability with the increase in the water content can be caused (Ng et al., 2020). That the permeability of sandy soil increases with the increase of moisture content found by van Verseveld and Gebert (2020) was mainly due to the formation of secondary macropores. It was likely due to the water-induced formation of soil aggregates when sand was compacted at higher water contents (van Verseveld and Gebert, 2020). The results indicate that soil pore size distribution can have significant effect on gas permeability. Possible larger diameter pores in soil can be resulted due to the change in water content in soil. Gas permeability of GCL is approximately 1–3 orders of magnitudes less than that of soil with the same volumetric water content (Fig. 4a). Permeability of GCL decrease significantly with the increasement of its water content. Permeability of gas through GCL range from 1



(a)



(b)

Fig. 4. (a) Permeability and (b) diffusion coefficient of cover soil obtained by column test from several review studies.

$\times 10^{-19}$ to 1×10^{-13} m^2, for a range of gravimetric moisture content (=10–150%) of GCL (Bouazza and Rahman, 2007; Rouf et al., 2016; Bouazza et al., 2017). However, when clay is under high compaction (>95%), the gas permeability of clay is similar with that of GCL (Didier et al., 2000; Ng et al., 2020).

Gas diffusion coefficients of soil of different tests show similar increasing trends with the increase of air-filled porosity (Fig. 4b). The tests shown in Fig. 4b were using oxygen except for the one conducted by Yilmaz et al. (2021). It can be concluded that moisture content, degree of compaction, the way of compaction, types of soil, and the additive (i.e., biochar) can have great impact on gas diffusion coefficient. The range of gas diffusion coefficient is 10^{-9}-10^{-5} m^2/s. The Millington and Quirk (1961) model was widely used for the calculation of gas diffusion coefficient. The dotted line shown in Fig. 4b was calculated based on the Millington and Quirk (1961) model for oxygen diffusion in clay soil with porosity equaling 0.489. It is indicated that the trends of Millington and Quirk (1961) model fits well with the laboratory results. However, it is indicated from Fig. 4b that for highly compacted soil (Hamamoto et al., 2011) and undisturbed landfill cover soil (Gebert et al., 2011), the model cannot give a good prediction. Macropores present in undisturbed soil will be retained. Thus, the soil structure is sustained. Hence, soil gas can move rapidly through the macropores, with the result that the diffusion coefficient of undisturbed soils would be larger than that of disturbed soils and the model predicted (Fujikawa and Miyazaki, 2005). For highly compacted soil, the quantity of connected pores is decreased. As a result, the diffusivity of gas is smaller than the Millington and Quirk (1961) model predicted. Besides, it is indicated that the Millington and Quirk (1961) model can give a good prediction of different textures of sieved and repacked soil (Jin and Jury, 1996).

Amendments of soil enhanced the performance of landfill covers due to improvement in the water retention, which led to the reduction of desiccation cracks formation and the emissions of LFG (Yargicoglu and Reddy, 2018; Raga et al., 2018). Many studies investigated the adsorption of methane and VOCs by biochar-modified soil to be used as landfill cover soil (Bushnaf et al., 2011; Sadasivam and Reddy, 2015b). The increased ratio of micropores in biochar makes it highly suitable for gas adsorption purposes (Brown et al., 2006). The pyrogenic production process of biochar increases its sorption potential when used as a soil amendment (Sadasivam and Reddy, 2015b). However, the impact of biochar modification on the permeability of gas can be neglected when the compaction of clay is less than 80% (Wong et al., 2016), which is mainly because biochar no longer acts as a filling material to retard gas flow (Sun et al., 2013). However, the soil cover is not free of cracks, which leads to break through of LFG when gas pressure increases. The pressure of gas break through increases directly with the thickness and the degree of saturation of the LFC. Compacted clay can impede gas breakthrough when the thickness of this layer is $\geq$ 0.6 m, and the degree of saturation is 60% for the case of low pressure ($\approx$10 kPa) (Ng et al., 2015a, 2015b). It was found that biochar modification of soil can increase the water retention capacity and decrease the crack intensity factor. Water retention capacities of 15%-biochar modified soil increased by 1.6 times compared with control soil without modification. The crack intensity factor was reduced from 7% to 2.8% for the same dose of biochar (Bordoloi et al., 2018).

The effects of temperature, wet-dry cycles, freeze-thaw cycles, landfill waste settlement on the permeability and diffusion of LFG through landfill covers, including soils, geosynthetics, and geomembrane, have not been investigated yet. The performance of GMBs and geosynthetics in eliminating emerging contaminants such as PFAS should also be considered for further studies (Bouazza, 2021).

3.2. Methane oxidation and VOC degradation of cover soil

Some CH_4 oxidation tests, including batch test, column test, and field test conducted recently, were listed in Table 3. Methane removal efficiency, which depends on the type of test, gas flow rate, environmental factors, and additive dose, was usually investigated by laboratory column tests (Yargicoglu and Reddy, 2017, 2018; Raga et al., 2018; Frasi et al., 2020; Huang et al., 2020; Niemczyk et al., 2022). It is revealed that methane removal efficiency of biochar-amended soil can reach more than 50%. Moisture distribution and infiltration of the cover can be affected greatly by the placement of a biochar layer (Yargicoglu and Reddy, 2017), which in turn significantly affects the methane oxidation (see Table 3). Relatively optimal soil moisture regime can be induced by the high-water retention capacity of biochar owing to its high surface area and internal porosity. Methane oxidation can also be enhanced by aeration due to increased oxygen entrance into soil. Apart from biochar, the efficiency of biowaste and compost modified soil in removing methane was also investigated (Raga et al., 2018; Niemczyk et al., 2022). Methane oxidation cannot always be improved by biocover such as compost modified soil because oxygen diffusion into soil is restricted due to high moisture content, exopolymeric substance and fine texture of material, which reduce effective CH_4 oxidation (Niemczyk et al., 2022).

Recently, studies have been performed on methane oxidation using alternative electron acceptors in deeper parts of landfill covers with the absence of oxygen. Batch and column tests were conducted by Parsaeifard et al. (2020) on anaerobic methane oxidation in the deeper parts of landfill covers. The results of the batch tests indicate soils modified with sulfate, nitrate, and the combination of sulfate and hematite show better methane removal efficiency as compared to control soil. The effect of methane generation inhibitor on net methane removal can be neglected. Greater methane removal rates can be observed with the increasing of initial methane concentration. The highest methane removal rate in the anoxic zone for soil amended with sulfate and iron combinedly was observed through column test, followed by soil amended with sulfate only. The test also demonstrated that methane was removed mainly by

Table 3
Some methane oxidation tests conducted in recent 5 years.

Material/soil type	Methane removal rates	Methane removal efficiency	Experiment conditions	References
15% (v/v) Biochar-amended landfill cover soil	NM	85.2%	75 cm high Column test	Huang et al. (2020)
15% (v/v) Biochar-amended landfill cover soil with daily active aeration	NM	90.6%		
a pilot-scale biocover (soil: perlite: earthworm cast: compost, 6:2:1:1, v/v)		35–45% in winter; 85% in spring; 86% in early summer and 96% in late summer	Field test with 0.5 m bio-cover packing materials	Lee et al. (2018)
a compost material subject to LFG diluted with atmospheric air resulting in CH4 concentrations of 5–10		98–100%	Column test	Thomasen et al. (2019)
Typical landfill cover soil	7.4 mol/m^2/day	55%		Park et al. (2008)
Typical landfill cover soil amended with earthworm cast and powdered activated carbon	14.6 mol/m^2/day	98%	1 m long column test	
soil amended with sulfate + hematite		10%	Anaerobic column test	Parsaeifard et al. (2020)
soil amended with nitrate		−5%		
Column test with a 2.5-cm 100% biochar layer		58%	1 m long column test	Yargicoglu and Reddy (2017)
Column test with 20-cm 2% biochar amended silty clay soil		63%		
Column test with 20-cm wood pellet amended silty clay soil		72%		
Soil unamended	107.53	93%		Yargicoglu and Reddy (2018)
2% biochar-amended soil applied at 20–40 cm depth	111.54	97%		
10% biochar-amended soil applied at 20–40 cm depth	111.75	97%		
10% biochar-amended soil applied at 0–60 cm depth	113.28	97%		
Sandy loam soil from top cover soil of a closed landfill		30.4%	Batch adsorption test	Huang et al. (2020)
2.7% biochar amended Sandy loam soil		46.4%		
Biochar-amended soils		70–90%	Field test	Yang et al. (2017)
Aged refuse		30–82%		
Inoculated farm soil		100%	Batch test	Syed et al. (2016)
Inoculated biochar		60–75%		

sulfate-reducing bacteria because hydrogen sulfide was measured in the headspace of these columns.

Various techniques such as immobilization and degradation have been used to minimize contaminant migration and emission (Höhener et al., 2006; Nikiema et al., 2007; Tassi et al., 2009; Biswas et al., 2015; Lakhouit et al., 2016). Therefore, understanding the mechanism of VOC degradation in landfill cover soil is of great importance. Biocover including sewage sludge modified waste char can be a potential alternative cover for VOCs degradation in landfills where VOCs removal efficiencies can be maintained >85% for the long-term (Scheutz and Kjeldsen, 2005; Tassi et al., 2009; Su et al., 2015; Qin et al., 2020). Petroleum-based hydrocarbons were found to be degraded by more than 200 species of bacteria, yeast and fungi (Adarsh, 2014). Adarsh (2014) summarized that degradation of BTEX (benzene, toluene, ethyl-benzene and xylene) in the vadose zone was first-order biodegradation. However, the biodegradation rate is still considerably uncertain. Scheutz and Kjeldsen (2005) found that the removal efficiency of chlorinated hydrocarbons is greater than 57% by degradation in soil cover systems at a landfill in Denmark based on active soil columns test. However, lower chlorinated compounds such as vinyl chloride and dichloromethane were degraded at the top of the column. Benzene and toluene can also be removed in the active column. It was found by Tassi et al. (2009) that the effect of microbial activity on VOCs is not obvious compared with the impact on methane. However, the behaviour of VOCs in the cover soil should also be considered apart from CO_2 and CH_4 emissions. It is found that terpenes, phenol, furans, and halogenated compounds cannot be influenced by degradation processes and only depend on waste composition (Randazzo et al., 2020). It is found by Scheutz and Kjeldsen (2004) that methane oxidation rate in soil decreased with the increasing concentrations of HCFC.

Overall, biochar- and compost-modified soils can help to improve the long-term capacity of landfill soils to degrade VOCs and oxidize methane, which may present an alternative to traditional cover amendments such as activated carbon. However, it should be realized that fugitive emission of CO_2 in the atmosphere is not desirable. Hence, Chetri et al. (2022) performed column studies to evaluate the effectiveness of the different combinations of covers by using basic oxygen furnace (BOF) slag and biochar-amended soil to mitigate simulated LFG containing CH_4 (48.25%), CO_2 (50%) and H_2S (1.75%) with an average flux rate of 130 g $CH_4m^{-2}d^{-1}$. They have found that column containing BOF slag layer and 5% (by weight) methanotrophic bacteria inoculated biochar-amended soil layer exhibited maximum capacity to remove LFG as compare to other amended columns. Furthermore, 5% activated biochar-amended soil showed the highest oxidation of CH_4 (143 g CH_4/g-day), whereas BOF slag exhibited maximum CO_2 removal (145 mg CO_2/g BOF slag). Chetri et al. (2022) also stated that BOF is a potential candidate for mineral carbonation, a common phenomenon for calcium-containing minerals, including free lime (CaO), portlandite [$Ca(OH)_2$] and larnite (Ca_2SiO_4), which can readily react with CO_2 to form stable carbonates, as well as iron oxides (FeO, Fe_2O_3). These byproducts react with H_2S forming iron sulfides.

$$\mathrm{MOH}\quad \text{or}\quad \mathrm{M'O} + \mathrm{CO_2} = \mathrm{M(NHCO_3)}\quad \text{or}\quad \mathrm{M'CO_3} \tag{15}$$

$$\mathrm{M^{++}} + \mathrm{H_2S} = \mathrm{MS} + 2\mathrm{H^+} \tag{16}$$

However, studies of the physical properties of the modified soil, such as the soil-water characteristic curve, mechanical property, water conductivity, etc., should be conducted further.

4. Modeling landfill gas transport through the cover system

4.1. Analytical models for LFG transport in LFC

Analytical models can provide a better understanding of contaminant transport mechanisms, which ultimately lead to a better prediction of the migration of contaminants. They can also be implemented to verify results from numerical modeling or field analysis (Nadarajah and Rowe, 1996; Feng and Zheng, 2015; Xie et al., 2016). Further, truncation errors and numerical dispersion can be avoided by using analytical solutions because they are generally derived from basic physical principles. In the analytical model, transport mechanism including diffusion, advection, adsorption and degradation are usually considered (Yao et al., 2015; Shi et al., 2016; Xie et al., 2016, 2017, 2018; Feng et al., 2018, 2019). Analytical models are usually developed by combining mass conservation equation with Fick's law and Darcy's law. Linear and equilibrium adsorption, constant environmental factors such as temperature and first-order degradation were usually assumed.

It is indicated from Table 4 that analytical models tend to consider a single LFG transport process involving mass transport or gas pressure variation. The coupled processes considering LFG mass transport of pressure variation combined with temperature variation, moisture transport, vegetation effect, cover settlement, LFG generation and collection were always not considered in the analytical models. Nevertheless, these processes are crucial and have a great impact on the performance of LFC systems, LFG migration, and emission (Lei et al., 2011; Ng et al., 2015a, 2015b; Manjunatha et al., 2020; Lu and Feng, 2020). Based on a coupled numerical model for water-gas-heat transport through unsaturated landfill cover soil, Ng et al. (2015a, 2015b) indicated that the coupled interactions between methane oxidation and water-gas-heat transfer should be incorporated. Heat and water generated by the oxidation of microorganisms should be considered in methane oxidation models when designing landfills. Otherwise, the difference in methane oxidation efficiency can reach up to 100%. It also indicates that when the soil water content is higher than the field capacity, methane oxidation efficiency can be decreased by microbial oxidation, which generates water. The reverse is possible when the water content is less than the field capacity. These coupled processes should be considered when developing new analytical models. Furthermore, some dimensionless design curves need to be developed for landfill cover system based on the analytical models. Numerical models tend to be used to solve the coupled processes of landfill gas transport, as discussed in the next section.

4.2. Numerical model for LFG transport in LFC

Compared with analytical model, the coupled equations combining water vapour, liquid water, surface water, heat transport, and energy balance can be developed by numerical model (Saito et al., 2006; Garg and Achari, 2010; Guan et al., 2016; Feng et al., 2017, 2017a; Bian et al., 2018; Lu et al., 2019, 2021). More complex processes involving phase change, gas phase dissolution in liquid, liquid phase evaporation and variation of leachate pH can be simulated (Lu et al., 2021). LFG are present as part of a multicomponent mixture of gases and tend to react with atmospheric gases to produce chemical species with different chemical and physical properties. Therefore, numerical modeling considering muti-gas transport in landfill was also investigated (Molins and Mayer, 2007; Binning et al., 2007; Zuo et al., 2020). The contribution of diffusion and advection to multi-component gas transport was usually the focus of current studies.

The following equation can be used to describe the one-dimensional diffusion-convection transport of multi-component gas mixture, which is a typical case for LFG, through a muti-layer (M layer) porous medium in landfills (Popov and Power, 1996).

$$n^j\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x}\left(D_i^j\frac{\partial C_i}{\partial x}\right) - \frac{\partial (C_i V^j)}{\partial x} + P_i^j - d_i^j C_i \tag{17}$$

($i = 1, \ldots, N\, j = 1, \ldots, M$).where C_i is the gas concentration of the ith gas (kg/m^3); D_{ij} is the gas diffusion coefficient of the ith gas in the jth layer (m^2/s); V_j is the advection velocity of mixture gas in the jth layer (m/s); P_{ij} is the production term for the ith gas in jth layer (kg/m^3/s); d_{ij} is the reaction constant for the ith gas in jth layer (1/s); and x is the distance.

Table 4
Coupled process in landfill gas migration for selected studies.

Target gas	Coupled process						Dimensions	Solution method	Reference
	Gas flow	Water flow	Heat transfer	Leachate transport	Waste or soil settlement	Waste degradation			
VOCs	✓						1D	Analytical solution	Xie et al. (2016, 2017)
Landfill gas	✓						2D	Analytical solution	Feng and Zheng (2015)
Landfill gas	✓					✓	1D	Analytical solution	Shi et al. (2016)
LFG	✓						1D	Analytical solution	Li et al. (2013)
Methane and oxygen	✓						1D	Analytical solution	Yao et al. (2015)
Methane	✓						1D	Analytical solution	Feng et al. (2018, 2019)
Methane	✓	✓	✓				1D	Finite Difference Method	Ng et al. (2015a, 2015b)
Vapour	✓	✓	✓				1D	Finite Difference Method	Saito et al. (2006)
Methane	✓	✓	✓				1D	Finite Difference Method	Garg and Achari (2010)
Methane	✓	✓		✓			3D	Finite Volume Method;	Feng et al. (2017)
Methane	✓	✓	✓				2D	Finite Difference Method	Feng et al. (2017a)
Landfill gas	✓	✓					1D	Finite Difference Method	Guan et al. (2016)
Methane	✓						1D	Finite Difference Method	Bian et al. (2018)
multi-component gas	✓						1D	Finite Difference Method	Zuo et al. (2020)
multi-component gas	✓						1D	Finite Difference Method	Binning et al. (2007)
Landfill gas	✓	✓			✓		3D	Finite Volume Method;	Lu et al. (2019)
Multi-component gas	✓	✓	✓	✓		✓		Finite Volume Method;	Lu et al. (2021)
Gas	✓	✓	✓				1D	Finite Difference Method	Bouazza et al. (2014)

Dusty-Gas Model can also be used to simulate multi-gas transport through porous media. The porous media can be viewed as (n+1)th component when considering n gas species transport through a porous system. The impact of the porous media was viewed as a "dusty gas" component in the Dusty Gas Model equations of the gas mixture. The model equation can be written as:

$$\sum_{\substack{j=1 \\ j\neq i,p}}^{v} \frac{X_i F_j^D - X_j F_i^D}{D_{ij}^*} - \frac{F_i^D}{D_i^K} = \frac{1}{kT} \nabla P_i \tag{18}$$

where X_i is the mole fraction of species i; F_i^D is the total molecular diffusive gas flux of species i. D_{ij}* is the effective molecular diffusivity in a porous medium; D_i^K is the effective Knudsen diffusivity of gas i in a porous medium; k is the Boltzmann constant; T is the absolute temperature; and P_i is the partial pressure of gas i.

The contribution of diffusion and advection to multi-component gas transport was usually the focus of current studies. Concerning the migration of multi-component LFG in landfill cover systems, more focus was paid to the interaction between different gas molecular including methane, oxygen, VOCs, and H_2S. The transport and interaction between oxygen, methane and carbon dioxide were usually investigated (Binning et al., 2007; Molins and Mayer, 2007; Zuo et al., 2020). The impact of van der Waals force, electrostatic interaction, collision, and chemical process between different species of gas molecule should also be considered in the analysis of multi-component LFG transport. The investigation among other LFG, such as VOCs, methane, oxygen, hydrogen sulfide, and ammonia should also be conducted to get a better understanding of the LFG transport mechanism. It is indicated from Table 4 that gas transport coupled with moisture transport and heat transfer were widely investigated in numerical simulation in landfill. Waste or soil settlement was comparatively rarely studied in gas migration. It may have great impact on solute transport or landfill gas generation and LFG transport in deforming covers (Yan et al., 2021, 2021a).

Landfill cover system tends to get fractures due to the wet-dry cycles, freezing and thawing cycle, differential settlement and so on (Sinnathamby et al., 2014; Chen et al., 2022). Chen et al. (2022) indicated that gas emission rate can be increased by 10 times with the presence of desiccation cracks when gas pressure equals 5 kPa based on a laboratory column test. The analytical and numerical solution of LFG transport in fractured cover system are scarce due to the anisotropy and heterogeneity of fractured cover. Some existing analytical solution of LFG transport through fractured cover is a simplified model with one regular-shaped fracture (Xie et al., 2019). The propagation of fracture induced by pressure and gas flow was not considered in the model. However, the existence of cracks in landfill cover provides a preferential gas flow path and will exacerbate odour nuisance and greenhouse effect. Further attention should be paid on gas transport in cracked LFC. Studies should also be conducted on the gas and pressure induced fracture in landfill cover system.

Although more comprehensive understanding of LFG transport can be obtained through numerical model, the validation of the coupled numerical results is still a tough task. Designed test is often controlled with only one variable while numerical model is coupled with many processes. In this case, a well-designed field and laboratory tests may be required. Besides, numerical models combined with test results can give more visualized and intensive insights.

5. Conclusions and perspectives

A comprehensive review of available studies on the experimental and numerical investigations on landfill gas transport and emission, which includes diffusion, advection, sorption, and degradation has been presented here with an intention to control LFG emissions from landfills. Conventional contaminants including VOCs, carbon dioxide, and methane emitted from landfill are widely investigated, whereas emission and transport of nitrous oxide and emerging pollutants, including PFAS in the LFCs have been overlooked in the previous studies. The effects of temperature, wet-dry cycles, freeze-thaw cycles, landfill waste settlement on the permeability and diffusion of LFG through landfill covers, including soils, geosynthetic clay liners, and geomembranes should be further investigated.

Methane emission concentration from LFC hotspots can reach up to more than ten thousand ppm, which is several orders of magnitude greater than the limit standard required by the environmental protection agency. Concentration of hydrogen sulfide emitted from landfills can be 2–5 orders of magnitude greater than the odour threshold, which would cause severe odour nuisance. Though the addition of additives and microorganisms facilitate an improved performance of emission reduction from LFC, the physical properties of the modified soil, such as the water retention characteristic, mechanical property, and water conductivity should be further investigated. The long-term performance of the amended cover soils should be assessed for field scale application. Analytical models can provide a better understanding of LFG transport mechanisms. They can also be implemented to verify results from numerical modeling. Analytical models have been developed for LFG transport in layered cover systems considering diffusion, advection and methane oxidation. The coupled effects of gas, water, and heat transport, and the vegetation effect need to be considered in developing new analytical models. Furthermore, the dimensionless design curves or simplified method should be developed for LFCs regarding gas transport on the basis of the analytical models. The LFG are a multicomponent mixture of gases and tend to react with atmospheric gases to produce chemical species with different chemical and physical properties. The coupled numerical model for multi-component gas transport should then be developed to simulate interactions between the gases under the coupled processes of gas, water and heat transport. At present, more attention has been paid on the interaction between different gas molecular including methane, oxygen, VOCs, and H_2S for numerical simulation. Furthermore, the effects of van der Waals force, electrostatic interaction, collision, and chemical process between different species of gas molecule should also be considered for multi-component gas transport in LFCs in the future.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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