1	Advancements in Municipal Solid Waste Landfill Cover System: A Review
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5	Jyoti K. Chetri*
6	Graduate Research Assistant, University of Illinois at Chicago, Department of Civil, Materials,
7	and Environmental Engineering, 842 West Taylor Street, Chicago, IL 60607, USA, e-mail:
8	jkc4@uic.edu (*Corresponding author)
9	
10	
11	
12	
13	Krishna R. Reddy
14	Professor, University of Illinois at Chicago, Department of Civil, Materials, and Environmental
15	Engineering, 842 West Taylor Street, Chicago, IL 60607, USA, e-mail: kreddy@uic.edu
16	
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## 24 ABSTRACT

Municipal solid waste (MSW) landfill cover systems have evolved from being merely a soil cap 25 to a multicomponent, nearly impermeable systems providing better control over infiltration and 26 landfill gas (LFG) emissions. Recently, there has been a widespread development of alternative 27 cover systems which addresses the shortcomings of conventional cover systems such as high 28 29 construction and maintenance costs, susceptibility to damage due to desiccation cracking and freezing, and ineffective control of LFG emissions. Landfills are regarded as the third largest 30 source of anthropogenic methane (CH<sub>4</sub>) emissions in the United States. Apart from CH<sub>4</sub>, landfills 31 are a significant source of various other gases such as carbon dioxide (CO<sub>2</sub>), hydrogen sulfide 32  $(H_2S)$  and several other odorous and non-methanogenic organic compounds (NMOCs). The 33 modern engineered landfills typically install gas collection systems in addition to the 34 conventional soil cover to mitigate LFG emissions. However, these systems are not always 100% 35 efficient in capturing all the emissions. Moreover, at the older landfills where installing gas 36 collection systems is not economical and practically feasible, the fugitive LFG emissions is a 37 persistent problem. In this regard, alternative cover systems with wide range of cover materials 38 have been explored to address the fugitive LFG emissions. This paper summarizes the 39 40 advancements in the MSW landfill cover systems over the years, along with the core mechanisms underlying their function. Then, advancements in the alternative cover systems, 41 42 including their advantages, are discussed. Finally, the research challenges/opportunities in the 43 field of exploring alternate landfill cover systems are presented.

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Keywords: Municipal solid waste; Landfill gas; Landfill odors, Alternative cover systems;
Biocover; Biogeochemical cover

## 47 **1 Introduction**

Thousands of tons of municipal solid waste (MSW) are generated annually across the globe, and 48 some of the major contributors are the high-income countries in North America, Europe, and 49 Central Asia (e.g., United States, Russia, Denmark, Switzerland, etc.) contributing about 34% of 50 the total waste generated in the world <sup>[1]</sup>. In the United States (US) alone, approximately 267.8 51 million tons of MSW was generated in the year 2017 of which nearly 52% was landfilled <sup>[2]</sup>, 52 making landfills an important part of the waste management system in the country. Landfills 53 have evolved from being mere open dumps to highly engineered and well-regulated waste 54 containment facilities. Resource Conservation and Recovery Act (RCRA) was passed in the US 55 to address the soaring volumes of municipal and industrial waste <sup>[3]</sup>. Since then, landfill 56 regulations have only been made stricter to limit the environmental pollution from landfill 57 leachate and landfill gas (LFG) emissions. 58

Modern engineered landfills are provided with nearly impermeable bottom liner and 59 cover systems, gas collection systems, and groundwater monitoring systems to minimize the 60 seepage of leachate and migration of gases into the atmosphere. Placement of waste in the 61 landfill is performed in various stages, and subsequently different types of covers are applied 62 (e.g., daily cover, intermediate cover, and final cover) to prevent exposure of waste to the 63 surrounding environment at different stages of landfill operation. At the end of the day, a layer of 64 65 soil (~150 to 300 mm thick) is placed over the daily placed and compacted waste as daily cover <sup>[4]</sup>. Various alternative materials other than soil such as shredded tires, wood chips, removable 66 textile cover or single use plastics are also used as daily cover materials as there are no 67 regulations regarding hydraulic conductivity to such covers <sup>[5]</sup>. Intermediate covers are applied at 68 69 those sections of the landfills where another lift of waste will not be placed within 60-90 days of

the waste placement. Like daily covers, there are no regulatory requirements governing hydraulic 70 conductivity of the intermediate covers <sup>[5, 6]</sup>. The final cover is placed when the landfill reaches 71 the designed waste capacity <sup>[7]</sup>. The primary function of the final cover system is to prevent 72 breeding of rodents and flies, ingress of precipitation into the waste, and migration of harmful 73 gases from the landfill into the atmosphere. The minimum regulations require the landfill cover 74 75 to have an infiltration layer and an erosion layer, however, landfill cover can have several layers depending on the site conditions, waste composition, and climatic conditions. The conventional 76 final cover systems typically have one or more barrier layers to restrict the infiltration and gas 77 migration<sup>[6]</sup>. 78

Over the years, various alternative cover systems such as evapotranspirative (ET) cover, 79 capillary barriers, anisotropic cover, and engineered turf cover have been developed as an 80 alternative to the conventional cover systems used in landfills. One of the major advantages of 81 the alternative cover systems is the reduction in construction and maintenance costs associated 82 with the conventional cover systems <sup>[6]</sup> and mitigation of damage due to physical and biological 83 processes which can further lead to increased infiltration <sup>[8]</sup>. Apart from infiltration issues, LFG 84 emission is another major issue of landfills. The increasing concerns regarding fugitive CH<sub>4</sub> gas 85 86 emissions from landfills has led to extensive research on the alternative cover materials which can mitigate the CH<sub>4</sub> emissions. In this regard, the CH<sub>4</sub> oxidation potential of the landfill cover 87 88 soils was explored extensively by various researchers and the studies related to alternative cover 89 systems have continually evolved to address other gaseous emissions in addition to CH<sub>4</sub>.

The main objective of this review is to outline the progressive development of the landfill cover systems over the years. The paper presents a comprehensive summary of the studies exploring alternative cover systems, their benefits, and challenges associated with them. The 93 study also reviews the type of studies (laboratory and field studies) performed to evaluate the 94 performance and efficiency of the different alternative cover systems. In addition, this paper 95 analyzes the underlying mechanisms that govern the functioning of different cover systems and 96 the parameters affecting their performance which can help to delineate the challenges for the 97 current and future research in the field of alternative cover systems for landfills.

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## 99 2 Cover Design Criteria

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As per the United States Environmental Protection Agency (USEPA), the specific functions of 101 landfill cover systems are to minimize vector breeding, control water movement to minimize 102 infiltration and erosion, control harmful gas movement, and minimize fire hazard potentials <sup>[9]</sup>. 103 The landfill cover regulations mainly focus on minimizing the infiltration into the waste until the 104 confined waste ceases to cause impermissible threat to human health and environment <sup>[10]</sup>. A 105 post-closure care period of 30 years is mandated for monitoring the integrity of the landfill 106 performance including the final cover system <sup>[11]</sup>. However, the design life of landfill cover may 107 be longer than the regulatory 30 years post-closure care period which depends on several factors 108 including service life of the materials used in cover construction <sup>[10]</sup>. Other than using durable 109 materials for cover construction, an adequate cover design involves ensuring the stability of the 110 veneer slope, sufficient internal drainage, surface-water runoff controls, surface erosion 111 protection, freeze-thaw protection, and ability to sustain sufficient vegetation <sup>[10]</sup>. Some of the 112 important aspects of cover design are explained in the following sections. 113

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115 Infiltration

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One of the primary functions of landfill cover system is to restrict the percolation of water into 117 the waste and minimize the amount of leachate generated in the landfill. Increased infiltration 118 leads to increase in leachate head at the landfill bottom liners thereby increasing the potential for 119 seepage which can possibly cause landfill slope failures and subsurface contamination. When the 120 precipitation exceeds runoff, evapotranspiration (ET) and any storage in the cover materials, the 121 water infiltrates into the waste. There are several water-balance models developed for wide range 122 of hydrologic problems. One of the water balance programs specifically developed for water 123 balance analysis of landfills, cover systems, and solid waste disposal and containment facilities is 124 Hydrologic Evaluation of Landfill Performance (HELP) model <sup>[12]</sup>. USEPA requires hazardous 125 and nonhazardous waste facilities to use HELP model to assess closure designs <sup>[13]</sup>. The model is 126 applicable for open, partially closed, and fully closed waste containment facilities. The model 127 takes into account various components of the landfill such as vegetation, cover soils, waste cells, 128 drainage layers, barrier layer, and geomembrane (GM) liners, and provides estimates for runoff, 129 evapotranspiration, drainage, leachate collection, and liner leakage <sup>[13]</sup>. Unsaturated soil water 130 and heat flow model (UNSAT-H<sup>[14]</sup>), HYDRUS <sup>[15]</sup>, and Finite Element subsurface FLOW 131 simulation system (FEFLOW<sup>[16]</sup>) are some of the computer programs used for simulating water 132 133 balance for landfill cover systems.

The RCRA regulations require landfills to provide ~45 cm (18 inches) thick barrier layer to minimize infiltration in an MSW landfill. However, in practice, a drainage layer made of granular soil or geosynthetic material (e.g., geotextile) is provided above the barrier layer or infiltration layer to intercept the infiltrating water and minimize percolation into the barrier layer and underlying waste. The drainage layer should have adequate flow capacity to minimize the
buildup of hydraulic head on the barrier layer <sup>[10]</sup>.

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## 141 Gas Emissions Control

The waste undergoes decomposition in different phases in the landfill and generates huge 142 amount of leachate and gases (mainly CH<sub>4</sub>, CO<sub>2</sub> and trace amounts of NMOCs. The gas 143 generation in the landfill depends upon waste composition, age of waste, presence of oxygen, 144 moisture content and temperature <sup>[17]</sup>. Various models have been developed to predict the gas 145 generation, gas composition, and spatial variability of gas generation in landfills, and are mostly 146 based on the zero, first or second order decay kinetics <sup>[7]</sup>. One of the most popular and simplified 147 mathematical tool for estimating LFG emissions from MSW landfills is Landfill Gas Emissions 148 Model (LandGEM) developed by USEPA. It is based on the first-order decay equation (Eq. 1) 149 and is used for quantifying annual LFG emissions over a period in an MSW landfill <sup>[18]</sup>. 150

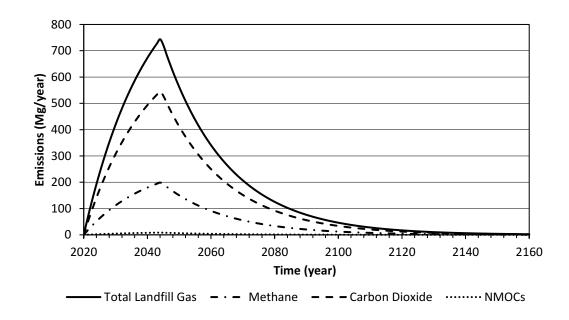
151 
$$Q_{CH_4} = \sum_{i=1}^{n} \sum_{j=0.1}^{1} k L_0 \left(\frac{M_i}{10}\right) e^{-kt_{ij}}$$
(1)

152 where,

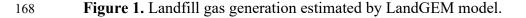
- 153  $Q_{CH_4}$  = annual CH<sub>4</sub> generation in the year of calculation (m<sup>3</sup>/year)
- i = 1 year time increment
- n = (year of the calculation) (initial year of waste acceptance)
- j = 0.1 year time increment
- 157  $k = CH_4$  generation rate (year<sup>-1</sup>)
- 158  $L_o = potential CH_4 generation capacity (m^3/Mg)$
- 159  $M_i = mass of the waste accepted in i<sup>th</sup> year (Mg)$

160  $t_{ij}$  = age of the j<sup>th</sup> section of waste mass M<sub>i</sub> disposed in the i<sup>th</sup> year (decimal years, e.g., 2.2 161 years)

The LandGEM model provides an estimate of the gas generation during the waste placement years, the corresponding emission rates, and an estimate for the waste stabilization period as well. **Fig. 1** shows the annual LFG emission rates for an MSW landfill with waste capacity of 60,702 megagrams (Mg) and annual waste input of 2,500 Mg/year. As shown in **Fig. 1**, LFG generation persists for a longer duration even after the closure of the landfill.



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The LFG migrates laterally and upwards through the landfill side walls and cover surface. Generally, landfill covers are placed to restrict the upward migration of the gases however, upward restriction leads to horizontal migration along the waste layers, ultimately making their way to the areas outside of the landfill <sup>[17]</sup>. Some of the major factors affecting the migration of gases in the landfills are diffusion, pressure gradient, permeability, and temperature <sup>[17, 19]</sup>. Diffusion is the movement of gases from areas of high concentration (within landfill) to the

regions with relatively lower concentration of the gases (e.g., atmosphere). The LFG is generated 175 in significant amounts and the gas movement is generally restricted by the compacted waste and 176 soil cover which leads to increase in gas pressure. This causes the advection of gases under the 177 pressure gradients. Specifically, the LFG tends to migrate through the path of least resistance. 178 Hence, they tend to migrate easily through coarse grained soils while the fine-grained soils like 179 clay offers more resistance to the flow. Temperature or heat generation during waste 180 decomposition also affects the migration of the gases <sup>[19]</sup>. Hence, the landfill cover should be 181 designed to control migration of LFG and prevent hazards associated with LFG. The federal 182 regulations (40 CFR part 60) require MSW landfills to install gas collection and control system 183 within 30 months after LFG emissions exceed a NMOC emissions rate of 34 Mg/year<sup>[20]</sup>. Active 184 and passive gas control systems are provided in modern landfills to control LFG emissions. The 185 passive systems divert the gas to a collection point or vent by natural pressure gradient 186 (advection mechanism), and active systems apply vacuum to channel the gas to the collection 187 point [6]. 188

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## 190 Slope Stability

Landfill covers are constructed with slight inclination to facilitate surface runoff and minimize ponding. Generally, the cover is designed to have a minimum slope of 2 to 5% at top deck as the slopes flatter than 2% may lead to ponding of water in the event of localized settlement <sup>[10]</sup>. However, steeper slopes are also not recommended as the potential for erosion and slope failure increases with increase in inclination <sup>[10]</sup>. The side slopes are made steeper with a typical slope of 2H:1V in case of soil cover, and 3H:1V or flatter in case of cover with geosynthetics <sup>[6]</sup>. Veneer slope failure is one of the commonly observed slope failures in landfills. Hence, landfill cover

should be designed to have sufficient stability during and after construction. Rigorous analysis of 198 slope stability should be performed by considering the shear strength of each component, 199 expected loading, and seepage pressures <sup>[21]</sup>. Similarly, the cover system incorporating 200 geosynthetics should be analyzed for interface slope stability as the stability is often impacted by 201 the interface shear strengths of the materials <sup>[6]</sup>. If the landfill is located in a seismic zone, the 202 203 landfill cover slope should be designed for seismic slope stability along with the static slope stability. Similarly, the MSW landfills generate gases in huge amount which may exert pressure 204 on the landfill cover thereby challenging their stability. Hence, the slopes should be stable 205 against the gas pressures that may develop in the cases when gas wells are not functioning 206 properly or are damaged or clogged due to perched leachate. 207

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- 209 **Runoff Control and Erosion Protection**

# Drainage and runoff control is a key aspect of landfill cover design. It is utmost important to minimize run-on into the active portion of the landfill as it may generate excess leachate. The typical runoff management strategies include construction of diversion berms, downslope flumes or channels, perimeter ditches, culverts, sedimentation, or detention basin. Diversion berms shorten the slopes, reduce erosions, and divert the runoff water. The downslope flumes carry runoff water from diversion berms to the perimeter diches through the side slopes. They should be designed carefully to accommodate the runoff velocities. The downslope channels are

susceptible to erosion from runoff and hence should be lined with riprap or reinforcement. Each element of storm water management system should be designed carefully as it may affect the overall stability of the landfill. The erosion potential varies according to climatic conditions. Covers in arid and semi-arid areas, and in steep slopes are more prone to erosion as these sites offer poor support for vegetation <sup>[10]</sup>. The cover should have sufficient vegetation as it substantially reduces the potential for surface erosion, reduces surface runoff velocity, and binds the soil strongly with root action. In the locations on the landfill (e.g., steep slopes) and for the landfills located in climatic conditions with higher erosion potential (e.g. arid and semi-arid regions), erosion control measures in the form of gravel, rip rap, or geosynthetic controls such as geogrids may be provided <sup>[10]</sup>.

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## 228 Durability

The current landfill regulations in the U.S. mandate a 30 year post-closure after care period, 229 however, the stabilization period for landfill waste may extend for over a hundred years which 230 calls for a requirement of longer design life of landfill components including landfill cover. A 231 properly functioning landfill cover is warranted for eliminating long-term post-closure leachate 232 and gas generation potential of MSW <sup>[21]</sup>. Landfill cover is subjected to wide range of climatic 233 conditions, and excessive settlements and subsidence. The post-closure total settlement may 234 range from 10 to 20% of the landfill height in an MSW landfill <sup>[21]</sup>. Hence, it is imperative to 235 consider long-term durability and integrity, and effectiveness in the design of landfill cover 236 237 system. Similarly, if geosynthetics like GM are part of the cover system, the geosynthetic material should be so chosen which can withstand excessive settlements, corrosive gases, heat, 238 and pressure, and require minimum post-closure maintenance. In addition, while designing 239 240 alternative cover systems, the material chosen should be stable for the long-term performance of desired function. For example, alternative cover systems such as biocovers are prone to self-241 degradation if the organic amendment contains unstable carbon thus hindering the CH<sub>4</sub> oxidation 242

potential of the biocover. Therefore, the integrity of the cover components is of utmostimportance in the selection and design of the cover system.

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## 246 Sustainability

Most of the cover systems are designed to limit infiltration and migration of gases, and little 247 regard is paid towards the sustainability of the cover materials and cover system as a whole. 248 Sustainability is often considered equivalent to environmental sustainability and any material 249 which does not engender harmful environmental impacts is considered environmentally 250 sustainable. However, sustainability is not just about environmental impacts. It is an 251 amalgamation of environmental, economic, and social aspects. A sustainable landfill cover 252 system is the one which is technically sound in executing the intended function while causing 253 minimum amount of net environmental, economic, and social impacts. For example, the use of 254 geosynthetics such as GM and geotextile (GTX) are gaining prominence in landfill cover design, 255 256 but it is not known how sustainable they are in terms of the environmental, economic and social impacts considering their entire life cycle stages (from material acquisition to their disposal). 257 Similarly, in alternative cover designs, the sustainability of the alternative cover components is 258 259 of prime importance. For example, waste materials such as sewage sludge are commonly used in biocovers for CH<sub>4</sub> mitigation. The waste materials may seem a sustainable choice for landfill 260 261 cover from economic point of view but its environmental (e.g., leaching toxic chemicals) and 262 social impacts (e.g., odors) need to be assessed before using it in the landfill cover. Therefore, sustainability assessment shall be incorporated in the design and development of the landfill 263 cover systems and the decision-making process. 264

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## 266 End Use of Landfill

Landfills are normally spread over many acres of land and the area often remains unused after 267 the closure of the landfill due to various health and environmental concerns. However, with the 268 stricter landfill regulations, and better leachate and gas management techniques, the end uses of 269 landfills are being explored extensively. There has been an increasing trend of real estate 270 development over the former landfill sites <sup>[22]</sup>. Similarly, the closed landfills can be used for 271 recreational parks and other land uses such as golf courses, playgrounds, ball fields, botanical 272 gardens, and residential development <sup>[23]</sup>. Landfill cover systems should be designed considering 273 the end uses of the landfill. For example, if the end use of the landfill is development of a 274 recreational park, then the cover should be designed to sustain vegetation and elevate the 275 aesthetics. Similarly, if the end use of the landfill is developing solar farm, then the landfill cover 276 can be designed as an exposed GM cover without a vegetative layer. There exist many 277 challenges in using landfill surface after closure some of which are subsidence, fugitive gas 278 emissions, and odor <sup>[23]</sup>. Hence, landfill cover should be designed to accommodate possible end 279 uses. 280

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#### 282 **Resiliency**

With the global climate change, the extreme climatic events are becoming a recurring event, and the impacts are being felt at every part of the world. For example, extreme precipitation, extreme drought, hurricane and storm surges, sea level rise, and saltwater intrusion are some of the commonly experienced extreme climatic events. Since landfills are essentially considered to be a storehouse for all the toxic pollutants, any kind of breach in the containment system may cause severe damage to the human health and environment. In this regard, the design of landfill cover

systems for resiliency is gaining wide prominence. Resiliency is the ability of a system to cope 289 up with the unforeseen changes in the environmental conditions without substantial damage and 290 quickly adapt to the changing conditions <sup>[24]</sup>. Given the magnitude of consequences associated 291 with failure of a landfill, it is imperative for the landfill to be resilient and be able to perform its 292 intended function in changing climatic conditions. Since the landfill cover is directly exposed to 293 294 the environmental conditions, it is important that the cover materials and the entire cover system itself are resilient to the changing environmental conditions. For example, the extreme flooding 295 events can jeopardize the functional performance of the landfill cover by increased infiltration in 296 conventional cover, water logging, and obstruction of gas transport in biocovers leading to 297 reduced CH<sub>4</sub> oxidation efficiency. These factors should be considered in the design of the cover 298 so that the performance of the cover system is not compromised by such extreme climatic events. 299

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## 301 **3 Regulatory Requirements**

In the U.S., MSW landfills are managed under RCRA Subtitle D [11]. Federal regulations 302 prescribed under 40 CFR Part 258, Subpart F, require that landfill owner/operators to place a 303 final cover system to reduce the infiltration of liquids and erosion of soil <sup>[11]</sup>. The permeability of 304 the final cover system should be less than the bottom liner system (if present) or the existing 305 natural subsoils and in no case, should it exceed  $1.0 \times 10^{-5}$  cm/s <sup>[6, 11]</sup>. As per the regulations, the 306 307 final cover should consist of an infiltration layer or barrier layer of a minimum of 45 cm (18 inches) of earthen material overlain by an erosion layer of a minimum of 15 cm (6 inches) of 308 earthen material capable of sustaining vegetation<sup>[3]</sup>. Fig. 2 shows the schematic of the Subtitle D 309 cover system for MSW landfills with and without GM liners at the bottom. 310

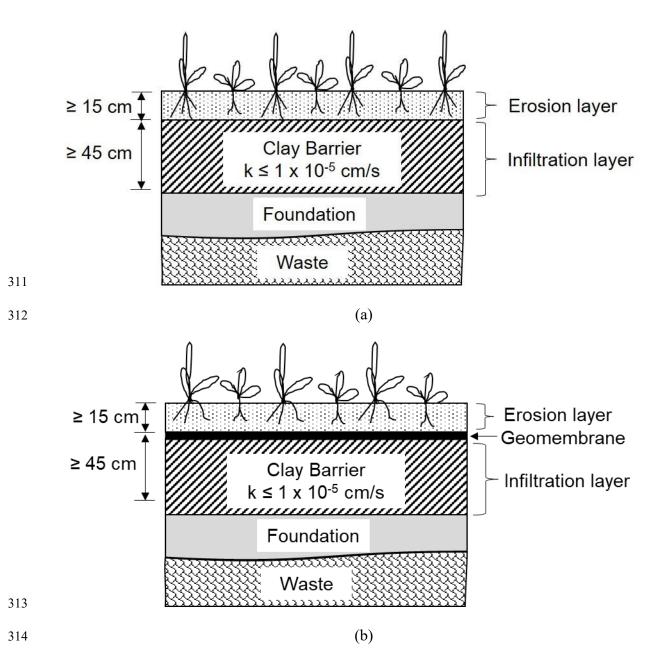


Figure 2. Subtitle D landfill cover system for a) unlined MSW landfill and b) MSW landfill with geomembrane liner at the bottom

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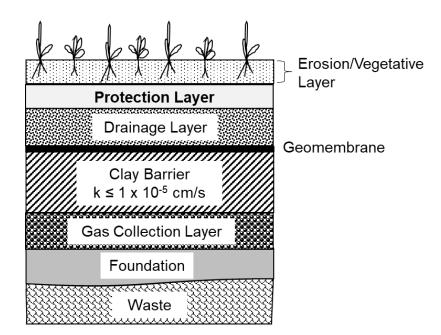
The regulations require barrier layer in the cover to have permeability less than or equal to that

- of the bottom liner, however, the use of GM in the cover is not obligated. If a GM is used in the
- bottom liner, then it becomes a necessity to use one in the cover to comply with the permeability

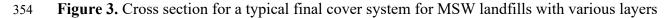
requirements. The landfill cover (the final cover) can have various components depending upon 321 the site conditions and anticipated gas generation. Fig. 3 shows a typical cross section of a 322 landfill cover with various components <sup>[25]</sup>. The top layer also called the vegetative or erosion 323 layer provides protection against erosion and supports vegetation if the climate supports 324 vegetation growth <sup>[4]</sup>. Sometimes, nutrients are added to the topsoil or the vegetative layer to 325 enhance vegetative growth <sup>[26]</sup>. An additional cover soil layer below erosion layer may be 326 provided as a protection layer in areas susceptible to frost degradation <sup>[25]</sup>. The protection layer 327 may serve to store excess infiltrated water which is removed later by ET<sup>[4]</sup>. Similarly, the areas 328 which receive substantial rainfall are provided with drainage layer to minimize seepage through 329 the barrier layer, reduce water head on the liner due to percolation, and reduce instability induced 330 by water pressure <sup>[4]</sup>. For the landfills where high CH<sub>4</sub> generation is anticipated, gas collection 331 layer is provided to install gas vents. Federal regulations allow the use of alternative cover 332 designs which can provide equivalent protection against infiltration and erosion; however, the 333 designs must be approved by authorized personnel<sup>[3]</sup>. 334

Different countries have different regulatory requirements. For example, in India, the 335 final cover is required to have a 60 cm thick barrier layer of clay or amended soil with 336 permeability less than  $1 \times 10^{-7}$  cm/s and an overlying 15 cm thick drainage layer. A 45 cm thick 337 vegetative layer shall be placed on top of the drainage layer to support natural vegetation and 338 protect from erosion <sup>[27]</sup>. Similarly, in Germany, the MSW landfills are grouped as Class I which 339 receive virtually inert waste with total organic carbon (TOC)  $\leq 20$  mg/L and Class II which 340 receive higher organic or degradable waste with TOC  $\leq 100 \text{ mg/L}^{[28]}$ . The Class I landfills are 341 also referred to as mineral solid waste landfills as they receive inert wastes which are not 342 expected to undergo chemical or biological reactions <sup>[29]</sup>. The Class II MSW landfills are 343

required to have a surface protection layer with thickness adequate for long-term protection. A 344 drainage layer of thickness 30 cm is provided below surface layer made up of granular soil with 345 minimum hydraulic conductivity of 0.1 cm/s. The drainage layer is underlain by hydraulic 346 barrier layer of 50 cm thickness made up of compacted clay with hydraulic conductivity  $\leq 5 \times 10^{-10}$ 347 <sup>7</sup> cm/s <sup>[29]</sup>. A high-density polyethylene (HDPE) GM of thickness  $\geq 2.5$  mm is placed over the 348 compacted clay barrier. A gas venting layer of thickness adequate to accommodate gas collection 349 pipes with minimum diameter of 100 mm is provided below barrier layer and finally a 350 foundation layer is placed above the waste to provide required gradation to the overlying cover 351 layers <sup>[29]</sup>. 352







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357 4 Alternative Cover Systems

As per the federal regulations, alternative cover systems which perform equivalent to a 358 conventional cover system can be used as MSW landfill cover. Alternative cover systems for 359 MSW landfills have been explored by various researchers from the past two decades. Cost is one 360 of the important considerations for exploring alternative landfill cover systems as the 361 construction and maintenance cost can be reduced significantly by using alternative covers at the 362 landfills <sup>[6, 30]</sup>. In addition to the cost considerations, the conventional cover system may not 363 always provide long-term protection against infiltration due to the formation of desiccation 364 cracks, limited water holding capacity of the topsoil, and increase in permeability of the barrier 365 soil due to freezing/thawing and root activity <sup>[8, 31]</sup>. In an assessment conducted in California, out 366 of 544 landfills in California which are located in wide variety of climatic conditions, 72-86% 367 were found to have failing compacted clay barrier and it was also found that the landfills, 368 irrespective of the climatic or geologic conditions, had failing clay barriers <sup>[31]</sup>. Hence, the need 369 to explore alternatives for conventional clay barrier was realized. Some of the alternative cover 370 371 systems are discussed in the sections below.

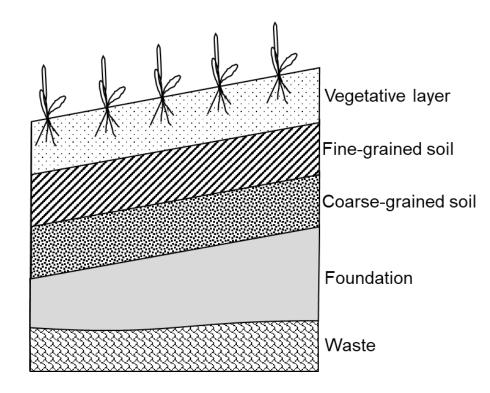
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## 373 4.1 Infiltration Cover Systems

374 *4.1.1 Capillary Barrier* 

A capillary barrier consists of a fine-grained soil layer underlain by a coarse-grained soil layer and the combination acts as a barrier for infiltrating water by capillary action <sup>[31-33]</sup>. A typical cross section of a capillary barrier cover system is shown in **Fig 4**. The cover system relies on the differences in the pore sizes of the fine-grained and coarse-grained soil layers for limiting infiltration through the cover <sup>[10]</sup>. When the water held in the fine pores of unsaturated soil meets the contrastingly larger pore sizes of coarse-grained soil layer, the capacity of the fine pores to

hold water at the existing matric suction reduces significantly. The water can advance only when 381 the matric suction is low enough to fill the pores with water i.e., at saturation <sup>[9]</sup>. In other words, 382 the integrated coarse-grained soil under a fine-grained soil layer system works on the principle of 383 contrasting hydraulic conductivities of the two soils at similar matric suctions <sup>[33]</sup>. Such type of 384 capillary barrier works effectively until the fine soil is fully saturated. Moisture accumulated in 385 the fine-grained soil layer needs to be removed to increase the efficiency of the cover which can 386 be done by evapotranspiration through the vegetative cover, or by lateral transport in an inclined 387 cover <sup>[33]</sup>. Morris and Stormont <sup>[33]</sup> compared the infiltration performance of basic capillary 388 barriers (0.6 m vegetative layer underlain by a coarse layer made up of gravel) and minimal 389 Subtitle D cover for five sites in the US using HELP and TRACER3D models. The results of 390 their study showed that an efficiently designed capillary barrier cover can perform equivalent or 391 superior to a minimal Subtitle D cover at many sites. They also evaluated the performance of the 392 capillary barrier with a transport layer at the interface of fine-grained and coarse-grained soil 393 layer and found a significant reduction in percolation. 394



**Figure 4.** Cross section of a typical capillary barrier cover system

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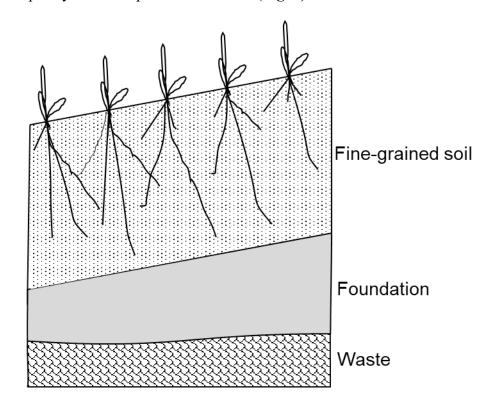
Over the years, many researchers have attempted to modify capillary barriers to serve for 398 purposes other than infiltration protection. One such example is the study by Berger et al.<sup>[34]</sup>, 399 who investigated CH<sub>4</sub> oxidation potential of capillary barrier comprised of compost-amended 400 401 sand overlying a layer of loamy sand. They reported CH<sub>4</sub> oxidation ranging from 57 to 98% in 402 the capillary barrier cover. In the recent years, studies have been conducted to modify capillary barriers to enhance the performance as well as the sustainability. For example, Rahardo et al.<sup>[35]</sup> 403 investigated the performance of dual capillary barrier using recycled asphalt pavement (RAP) 404 waste materials as the fine- and coarse-grained soil as an alternative to natural soil. The dual 405 capillary barrier comprised of two composite layers of each fine-grained RAP material overlying 406 a coarse-grained RAP material layer. Seepage analysis was performed using Seep/W software 407 after establishing saturated and unsaturated properties through laboratory testing. The dual 408

capillary barrier using RAP waste material showed efficacy in preventing rainwater infiltration 409 and hence was found to be a sustainable alternative to natural soil or aggregates. Similarly, Ng et 410 al. <sup>[36]</sup> investigated a modified capillary barrier by adding a fine-grained soil (clay) layer beneath 411 the two-layered capillary barrier layer (silt layer overlying gravel layer). They carried out column 412 tests simulating one-dimensional (1D) water infiltration and performed transient seepage 413 414 simulations to simulate the performance of three-layered modified capillary barrier cover. The results from their study indicated that the addition of clay layer at the bottom of two-layer barrier 415 system enhances the percolation protection significantly making it effective for a rainfall return 416 period of more than 1,000 years. However, the performance of such cover systems needs to be 417 verified through extensive field studies. Capillary barriers can serve as an effective landfill cover 418 barrier component mainly in the arid and semi-arid climates <sup>[33, 37]</sup>. Nevertheless, the 419 performance of the capillary barrier cover system may be limited in the regions which receive 420 heavy rainfall annually. One important aspect that needs attention in using capillary barriers is 421 the propensity of occlusion of pores in fine grained soil upon saturation leading to obstruction of 422 gas transport which may lead to accumulation of landfill gas underneath the barrier layer if there 423 are no provisions for gas management such as gas wells or gas collection headers in the cover. 424

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426 4.1.2 Evapotranspirative Cover System

Evapotranspirative (ET) cover is an alternative cover system which utilizes natural processes to protect infiltration of water into the waste <sup>[8]</sup>. Two major phenomena are used in ET covers to minimize infiltration: 1) water retention by soil making it available for plants and 2) evapotranspiration from soil and plants removing water from the soil <sup>[8, 38-41]</sup>. The ET cover works on the principle of water balance and functions based on the soil properties such as soil texture and water holding capacity to store water <sup>[40, 42]</sup>. ET covers are also referred to as water
balance covers and are mostly preferred over conventional cover in semiarid and arid climates
<sup>[38-40, 42]</sup>. The ET covers are designed as monolithic cover with a single fine-grained soil layer to
absorb water and bear vegetation (Fig. 5) or modified by adding a coarse-grained soil underneath
to form a capillary barrier explained heretofore (Fig. 4).



**Figure 5.** Cross section of a typical Evapotranspirative cover system

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The major advantages of ET covers are: 1) they are less prone to failure through desiccation, cracking and freeze/thaw cycles, and 2) require lower cost for construction and maintenance than conventional covers. The ET covers do not have compacted barrier layer which saves a fair amount of cost in labor and equipment. Locally available soils are typically used for ET covers averting the purchase or supply of clay soil. The operation and maintenance cost for ET covers are also lower than the conventional covers <sup>[8, 42]</sup>. ET cover designs are

affected by local soil type and resources, evapotranspiration potential, native plants, and the interactions of plants with soil and the resulting water balance <sup>[8]</sup>. The thickness of the ET covers is designed to store the water for the most critical climatic events. Soil layers of 0.60 m to 3.0 m thickness have been used in monolithic ET covers <sup>[42]</sup>.

In 1998, USEPA initiated a program called Alternative Cover Assessment Program 450 (ACAP) to obtain field-scale performance data for alternative covers <sup>[43,44]</sup>. Test facilities were 451 established at twelve sites across the US with broad sampling of the environmental factors which 452 affect the performance of landfill cover system <sup>[43]</sup>. Abichou et al. <sup>[44]</sup> assessed the performance 453 of ET cover in relation to a conventional cover system at one of the ACAP test sites by 454 monitoring percolation rates through the covers. The ET cover comprised of a 0.7 m thick 455 compacted soil overlain by a 0.6 m thick 3:1 mixture of soil and peanut hull compost. The ET 456 cover was vegetated with hybrid poplar trees and underwood of bermudagrass. The results 457 showed that the use of ET cover reduced percolation by 43% in comparison to conventional 458 cover. Similarly, Barnswell and Dwyer<sup>[45]</sup> assessed the long-term performance of ET covers for 459 MSW landfills in Northwestern Ohio. The ET cover was designed to generate percolation rates 460 less than 32 cm/year which is accepted by Ohio Environmental Protection Agency (OEPA) using 461 462 dredged sediment amended with sewage and lime sludge. The ET covers were constructed in drainage lysimeter and simulated 100-year rainfall events. The percolation rates through ET in a 463 464 one-year monitoring period were much lower than the OEPA standards. The mature plants were found to have better water balance than immature plants. 465

Although ET covers have been a popular alternative to conventional covers due to the lower cost, self-renewing and aesthetic qualities, they have certain limitations which include typical applicability in arid and semiarid climates, effectiveness is affected by local climatic 469 conditions such as precipitation, snowpack, etc. Similarly, the ET performance is significantly 470 affected by the vegetation type and the time duration when the vegetation is not mature. 471 Vegetation plays a major role in ET process; hence the design of the ET covers should consider 472 growth period of vegetation and potential saturation of the ET layer in the event of any heavy 473 precipitation before full development of vegetative layer which may hamper the proposed 474 function of the cover system.

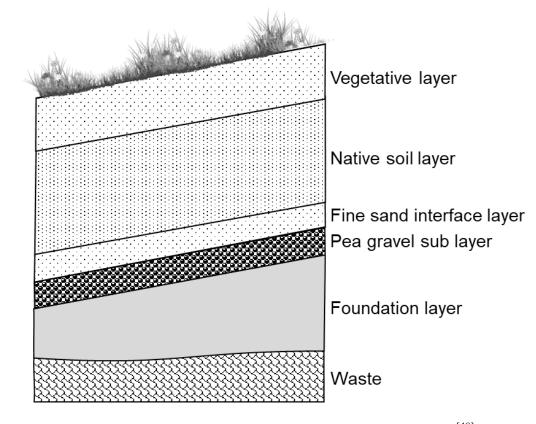
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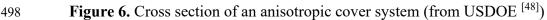
## 476 4.1.3 Anisotropic Barrier

Anisotropic barrier is a type of capillary barrier which is constructed by layering of capillary barriers. They are designed to restrict the downward flow of water and simultaneously stimulate the lateral flow of water <sup>[46-48]</sup>. The cover comprises of layers with variation in soil properties and compaction techniques to enhance the capillary forces and render anisotropic properties to the cover system <sup>[48]</sup>.

The US Department of Energy (USDOE) with an initiative of improvement of environmental 482 restoration and management technologies started the Alternative Landfill Cover Demonstration 483 (ALCD) program. It involved a large-scale field demonstration of performance of different 484 conventional and alternative covers at Sandia National Laboratories in Albuquerque, New 485 Mexico <sup>[46]</sup>. Two conventional cover designs (RCRA Subtitle D and Subtitle C covers) and four 486 alternative covers (ET, capillary barrier, geosynthetic clay liner (GCL) cover, and anisotropic 487 barrier) were constructed side by side. The cover performance was evaluated based on the flux 488 rates (percolation, mm/yr) and efficiency (percolation/precipitation\*100). The anisotropic cover 489 had four layers (from bottom to top): 15 cm thick gravel drainage layer, 15 cm thick fine-grained 490 sand interface layer, 60 cm thick non compacted native soil layer and a 15 cm thick top 491

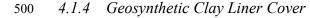
492 vegetation layer comprised of local topsoil and pea gravel <sup>[48]</sup> (Fig. 6). The anisotropic barrier, 493 ET cover and Subtitle C cover performed significantly well whereas the Subtitle D cover did not 494 perform well during the five-year testing period <sup>[49]</sup>. Anisotropic barrier and ET covers are much 495 cheaper to install than the subtitle C cover <sup>[46]</sup>. Anisotropic barrier covers perform better in arid 496 and semiarid climates; however they also suffer from similar limitations as ET covers.





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501 Geosynthetic clay liners (GCLs) were developed as a synthetic replacement to the conventional 502 compacted clay liners. GCLs are the hydraulic barriers which consist of clay (mainly bentonite) 503 either sandwiched between GTX or bonded to GMs in some cases <sup>[50-52]</sup>. GCLs prove more cost-504 effective in regions where low permeability clay is not locally available, and the thinner structure

of GCL reduces the space requirement while increasing the landfill waste capacity <sup>[53]</sup>. GCLs 505 gained prominence in barrier applications in landfills due to their low hydraulic conductivity 506  $(\sim 10^{-9} \text{ cm/s})$ , ease of installation, lower thickness, lower cost, and resiliency to adverse 507 environmental conditions such as freeze-thaw and wet-dry cycles <sup>[50, 52, 54]</sup>. The lower hydraulic 508 conductivity of GCLs is attributed to the physicochemical properties of bentonite which is a 509 510 naturally occurring clay with characteristic high swelling potential, ion exchange capacity, and low hydraulic conductivity <sup>[55]</sup>. Most of the commercial GCLs use sodium (Na) bentonite due to 511 their low hydraulic conductivity ranging from  $6 \times 10^{-10}$  to  $2 \times 10^{-9}$  cm/s <sup>[50, 52, 56,57]</sup>. Na bentonite 512 is composed primarily of the mineral Na montmorillonite which has high surface area, high 513 cation exchange capacity, and ability for interlayer swelling which contributes to the high 514 swelling potential and thus low hydraulic conductivity <sup>[54]</sup>. The water, when comes in contact 515 with the Na bentonite, is bound to the clay mineral surface, also called swelling, thereby sealing 516 off the macroscopic flow paths and increasing the tortuosity of the flow paths <sup>[54, 56]</sup>. The volume 517 of interlayer bound water is associated with the degree of swelling and hydraulic conductivity of 518 the bentonite in the GCL <sup>[56]</sup>. Swelling properties of bentonite renders unique self-healing 519 abilities to the holes and cracks formed during the operation of the GCL as a barrier <sup>[53]</sup>. It has 520 been reported that a hole as large as 75 mm in diameter can self-repair maintaining the original 521 properties which makes GCL a perfect candidate for barriers in landfill applications <sup>[53]</sup>. GCL, 522 especially with needle punched or stitch bonded GTX, can provide appreciable shear strength 523 with high internal shear resistance. Koerner et al. <sup>[58]</sup> assessed the internal shear strength of the 524 GCLs in landfill cover in fourteen full scale test plots. The test plots involved two cover designs: 525 one with GCL alone with 0.3 m sand drain layer and 0.6 m cover soil layer overlying the GCL, 526 527 and the second cover design had a GCL/GM composite (GCL beneath GM) as a barrier layer. A

geocomposite drainage layer made of geotextile/geonet/geotextile combination was overlain on a 528 GCL/GM combination and a cover soil layer of 0.9 m was placed above the geocomposite 529 drainage layer. In the 24 months of operation, only two slides and one internal slide were 530 reported. The interface slide was between GM and GTX interface which was attributed to the 531 extrusion of the bentonite through the GTX and lubricating the interface. The internal slide was 532 attributed to the installation inconsistencies rather than GCL functioning. Overall, the GCLs 533 showed appreciable internal shear resistance, low differential deformations, and better slope 534 stability. 535

Since the introduction of GCLs in 1986 <sup>[53]</sup>, extensive research has been conducted to 536 assess the performance of GCLs as a barrier material. Studies have shown that several factors 537 such as ion exchange, desiccation, penetration of roots from vegetation, humidity, confining 538 pressure, and age of installation can alter the hydraulic conductivity of GCL barriers significantly 539 <sup>[50-52, 59]</sup>. An increase in hydraulic conductivities of GCLs by an order over 5 was observed by 540 Meer and Benson <sup>[52]</sup> while analyzing the GCLs exhumed from four landfills after 4.1 to 11.1 541 years of installation. A complete exchange of Na with calcium (Ca) and magnesium (Mg) was 542 observed with swell index similar to Ca or Mg bentonite. It was concluded that the cation 543 544 exchange is inevitable unless the underlying or overlying soil is rich in Na. In addition, desiccation combined with cation exchange can lead to irreversible increases in hydraulic 545 conductivity of the GCLs. Melchoir et al. <sup>[60]</sup> presented the results of the long-term (18 years) 546 performance of different landfill cover systems, which included cover system with GCL barrier, 547 equipped with in situ large scale lysimeters at a landfill in Germany. The GCL which was 548 covered by 0.15 m of gravel layer and 0.30 m of cover soil showed significant crack formations 549 550 and seepage after within three years of operation. Exchange of Na ions with Ca and Mg ions

were observed. Mackey and Olsta <sup>[51]</sup> who analyzed the performance of GCLs used in landfill covers in two landfills in Florida also had similar observations. They suggested that providing a thicker soil cover above GCL or a GM can protect the GCL from desiccation and root penetration which may help to minimize increase in hydraulic conductivities despite ion exchange. The observations from various studies suggest that GCLs should be used with precaution in landfill cover applications.

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## 558 4.1.5 Exposed Geomembrane Cover

Exposed geomembrane cover (EGC) has been used in many landfills in place of conventional 559 cover systems. The EGCs do not incorporate overlying drainage layer and topsoil or erosion 560 layer provided in a typical landfill cover system <sup>[61]</sup>. In a typical conventional cover with GM, it 561 is covered with soil layer to support vegetation as well as reduce direct damage to the GM. 562 However, such cover systems are susceptible to slope failure due to slippage at interface <sup>[62, 63]</sup>. 563 The EGCs are preferably provided for the interim or temporary cover applications <sup>[62-64]</sup>. The 564 major benefits of providing EGC in intermediate cover are significant reduction in the amount of 565 percolation of precipitation and containment of LFG<sup>[63]</sup>. Reduced percolation results in reduced 566 leachate generation and thus significant cost reduction in leachate management. Providing EGC 567 in interim cover also helps to protect steep slopes with no potential erosion which is otherwise a 568 bigger concern for soil covers. Apart from interim cover, EGC can also be placed as a final cover 569 in the landfills, however it will require stricter considerations for long-term stability and thus 570 may call for the use of high-interface friction GMs and management of surface water drainage 571 [64] 572

EGC have been used in many landfill projects across the US. Although EGC application 573 was approved for interim cover since 1992, the use as final cover was approved much later <sup>[65]</sup>. 574 For example, Sabine Parish Landfill, Louisiana was permitted to use EGC as final cover in 1999 575 and the performance was encouraging [66, 67]. Similarly, Yolo County landfill near Davis, CA 576 chose to use EGC for the bioreactor landfill <sup>[68, 69]</sup>. The anchor trenches, provided to hold GM in 577 place, were backfilled with soil and temporary ballast consisting of 20 kg sandbags were 578 installed to counteract the uplift forces caused by the wind and protect the exposed GM <sup>[68]</sup>. 579 Hickory Ridge landfill, Altanta, GA is another landfill to use EGC as final cover and was the 580 first fully permitted EGC final cover closure system <sup>[70]</sup>. The 48-acre landfill used 1.5 mm thick 581 Thermoplastic Polyolefin (TPO) reinforced GM for EGC. The EGC covered landfill was 582 combined with solar cap technology by installing solar panels over the EGC cap and was 583 transformed into the biggest solar energy generating facility in Georgia as well as became the 584 world's largest solar energy cap. 585

Although, EGC provides cost saving in terms of construction and maintenance, it is 586 subjected to degradation through various mechanisms other than accidental damages. The 587 lifetime of the GMs reduces significantly in the exposed condition due to the major degradation 588 mechanisms; ultraviolet radiation, elevated temperatures, and atmospheric oxidation <sup>[71]</sup>. Where a 589 nonexposed HDPE GM can have a lifetime of 166 to 446 years between temperatures of 30 to 20 590 °C, for an exposed HDPE GM the lifetime can be as much as 36 years <sup>[72]</sup>. In addition, EGCs are 591 exposed to high uplift pressures from wind which requires installation of numerous anchor 592 trenches as well as access to the EGCs during post closure care can be difficult <sup>[73]</sup>. 593

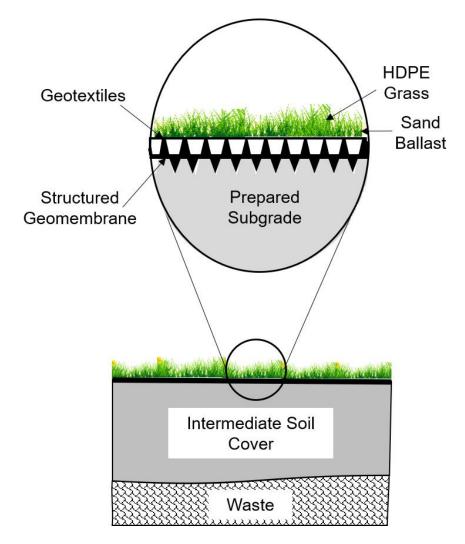
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## 595 4.1.6 Engineered Turf Cover

In the recent years, attempts have been made to overcome the shortcomings of the EGC by 596 introducing engineered synthetic turf cover. One of the patented turf covers comprise of 597 synthetic grass and GM<sup>[74]</sup>. The synthetic grass comprises of GTX clumped with synthetic 598 strands manifesting the appearance of grass. The GM used is usually textured GM liner, however 599 600 it can be designed as a drainage liner comprising of GM with geonet drainage media, or a drain liner with studs integrated with HDPE sheet <sup>[74]</sup>. Such type of cover system not only reduces 601 construction cost by eliminating the need for cover soil and reduces operation and maintenance 602 requirement annually, but also enhances the aesthetic appearance of the landfill. 603

Several configurations of turf covers have been explored to enhance their performance in 604 the dynamic environmental conditions. West et al. <sup>[73]</sup> presented an engineered turf cover for 605 final landfill cover which comprised of synthetic turf and impermeable GM layer (Fig. 7). The 606 synthetic turf comprises of UV resistant polyethylene turf with sand infill which provides 607 additional protection and increases longevity of the underlying GM. The GM used by West et al. 608 <sup>[73]</sup> was textured, structured GM with drainage studs and downward spikes which provide high 609 friction angles, resistance against sliding failure, and facilitates drainage under various gradient 610 611 conditions. The impermeable GM layer can sustain high vacuum pressures applied by active gas collection systems at landfills. Sanchez and Zhu [75] performed a comparative analysis of 612 stormwater pond design between conventional cover system and engineered turf cover system 613 also known as ClosureTurf cover which had similar configuration as West et al. <sup>[73]</sup>. Their 614 analysis results showed that the engineered turf cover system generates higher surface runoff 615 than a conventional cover system which engenders the need for a slightly deeper perimeter 616

drainage channel and larger stormwater pond. Simultaneously, it reduces the infiltration byincreasing the runoff.



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**Figure 7.** Schematic of turf cover system (modified from West et al. <sup>[73]</sup>)

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Engineered turf covers are increasingly being adopted by the landfill operators due to the ease of installation, applicability in steep slopes, and reduced construction cost, and reduced operation and maintenance requirements. Some of the completed projects include Bi County landfill, Tennessee, Berkeley County landfill, South Carolina, and Hartford landfill, Connecticut. Engineered turf covers have been successfully implemented in many states across the US and

have been considered a remarkable innovation to enhance the aesthetics of the landfills. However, the long-term performance of such type of cover systems needs to be validated with extensive field studies. In addition, the availability of such cover systems in developing countries or low-income countries could be a concern.

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## 632 4.2 Gas Mitigating Covers

Initially, the regulatory landfill cover designs were solely based on the infiltration consideration 633 with the focus on reducing leachate generation and preventing CH<sub>4</sub> generation to prohibit the 634 incidences of landfill fires or explosions <sup>[76]</sup>. LFG is typically composed of nearly 50% ( $\nu/\nu$ ) 635 CH<sub>4</sub>, 50% (v/v) CO<sub>2</sub> and trace NMOCs <sup>[11]</sup>. CH<sub>4</sub> and CO<sub>2</sub> constitute 16% and 65%, respectively 636 of the global anthropogenic (GHG) emissions <sup>[77]</sup>. CH<sub>4</sub> is a more potent GHG than CO<sub>2</sub> with 637 global warming potential (GWP) of 28-36 over 100 years <sup>[78]</sup>. CH<sub>4</sub> is a short-lived gas with 638 atmospheric lifetime of 12 years however, its radiative energy is much higher than CO<sub>2</sub> <sup>[7, 79]</sup>. 639 Atmospheric CH<sub>4</sub> concentration has increased tremendously over the years and surpassed the 640 pre-industrial period by 150% [80]. MSW landfills are the third largest source of anthropogenic 641 CH<sub>4</sub> emissions globally as well as in the US and accounted for 17.4 percent of total CH<sub>4</sub> 642 emissions in the US in 2018 <sup>[78, 81]</sup>. Because CH<sub>4</sub> has more potential to trap heat than CO<sub>2</sub> and 643 landfill serves as a major source of anthropogenic CH<sub>4</sub> emissions, continuous efforts have been 644 made towards controlling landfill CH<sub>4</sub> emissions. 645

The emission guidelines for MSW landfills (40 CFR 60) require landfills to install gas collection and control system for open landfills if NMOC emissions exceed 34 Mg/year or the surface CH<sub>4</sub> emissions exceed 500 parts per million (ppm), and for closed landfills if NMOC emissions exceed 50 Mg/year <sup>[20]</sup>. Thus, gas collection systems are installed at landfills for LFG

emissions control as well as beneficial use of CH<sub>4</sub>. However, gas collection is efficient only after 650 the placement of impermeable final cover which often takes several years from the start of waste 651 disposal point <sup>[82]</sup>. Typically, intermediate covers are installed before placement of final cover 652 however, they are not impermeable to gas migration. Besides, the installed gas collection 653 systems may not be 100 percent efficient in capturing all the gases generated in the waste. LFG 654 655 collection efficiency varying from 50% to 100% (average 75%) depending on the cover type and coverage of the collection system has been reported <sup>[83, 84]</sup>. Hence, a fraction of LFG is often 656 emitted into the atmosphere despite having gas collection system in place. Moreover, at the older 657 and abandoned landfills where providing gas collection system is not economical or practically 658 feasible, the problems of fugitive emissions is preeminent. As a result, in the recent years, focus 659 has been shifted to developing alternative cover systems which can mitigate fugitive CH4 660 emissions from landfills. 661

Bogner et al. [85] monitored landfill emissions at two landfills located at two different 662 climatic regions: Illinois (Mallard Lake) and California (Brea-Olinda) using a closed chamber 663 technique. During the monitoring period of 1988-1994, landfill cover soil was found to act as a 664 sink for CH<sub>4</sub> in well-aerated regions and the consumption of CH<sub>4</sub> was attributed to CH<sub>4</sub> 665 666 oxidation. CH<sub>4</sub> oxidation by microbes was identified in early 1900s and the first CH<sub>4</sub> oxidizing bacterium was isolated by Söhnhen in 1906 [86]. Earlier, the CH<sub>4</sub> consumption in aerated 667 temperate forest soils had been reported <sup>[87, 88]</sup>. Apart from forest soils, CH<sub>4</sub> oxidation potential 668 669 has also been reported for wide range of natural environments such as agricultural soils, wetlands, rice paddy fields, and peatlands <sup>[76, 89]</sup>. Whalen et al. <sup>[90]</sup> investigated the CH<sub>4</sub> oxidation 670 potential of topsoil from a park constructed over a landfill in California. They observed high CH4 671 oxidation rates (45 g m<sup>-2</sup> d<sup>-1</sup>), which was the highest reported value for any environment to that 672

date. Likewise, Kightley et al. [91] performed laboratory incubations of different soils with CH4 673 for six months and evaluated their CH<sub>4</sub> oxidation potential. In their study, porous coarse sand 674 showed highest CH<sub>4</sub> oxidation potential (166.5 g m<sup>-2</sup> d<sup>-1</sup>) among all the soils tested. Similarly, 675 Bogner et al.<sup>[92]</sup> evaluated CH<sub>4</sub> oxidation rates in landfill cover soil at Northeastern Illinois 676 landfill using static flux chamber technique under real field conditions. They observed a swift 677 change in CH<sub>4</sub> oxidation rates (up to 4 orders of magnitude) with change in CH<sub>4</sub> concentrations. 678 Negative fluxes of CH<sub>4</sub> were observed at locations near and far from the gas collection wells, 679 even into full winter with freezing conditions, showing high CH<sub>4</sub> oxidation potential in soil and a 680 maximum oxidation rate of 48 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> was observed. In another study, Scheutz et al. <sup>[89]</sup> 681 investigated the attenuation of CH<sub>4</sub> in landfill cover soil sampled from a location emitting CH<sub>4</sub> 682 by performing soil microcosms incubation and observed a high rate of CH<sub>4</sub> oxidation ranging 683 from 24 to 112 µg CH<sub>4</sub> g<sup>-1</sup> h<sup>-1</sup>. Likewise, Scheutz and Kjeldsen <sup>[93]</sup> performed batch incubation 684 experiments on landfill cover soil obtained from a depth of 15 to 20 cm below ground surface 685 (bgs) at room temperature (22 °C) and attained a maximum oxidation rate of 104 µg CH<sub>4</sub> g<sup>-1</sup> h<sup>-1</sup>. 686 In a similar laboratory incubation experiment by Reddy et al. <sup>[94]</sup>, a maximum CH<sub>4</sub> oxidation rate 687 of 195 µg CH<sub>4</sub> g<sup>-1</sup> h<sup>-1</sup> was attained at 30 °C in landfill cover soil sampled from a depth of 30-60 688 cm bgs. 689

690 CH<sub>4</sub> oxidation occurs in landfill cover soils with the help of methanotrophic bacteria. The 691 continuous influx of CH<sub>4</sub> from the underlying waste results in the enrichment of cover soil with 692 CH<sub>4</sub> oxidizing bacteria called methanotrophs. CH<sub>4</sub> serves as the sole source of carbon and energy 693 for the methanotrophs. Methanotrophs oxidize CH<sub>4</sub> to CO<sub>2</sub> in the presence of oxygen as shown 694 in **Eq. 2** <sup>[7, 95]</sup>.

695  $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$  (2)

The complete methanotrophic CH<sub>4</sub> oxidation process involves oxidation of CH<sub>4</sub> to methanol, 696 methanol to formaldehyde, formaldehyde to formate and finally to CO2 [86, 96-97]. The 697 methanotrophic CH<sub>4</sub> oxidation is catalyzed by various enzyme and enzyme methane 698 monooxygenase (MMO) is a key enzyme to catalyze oxidation of CH<sub>4</sub> to methanol <sup>[86, 93, 95-96]</sup>. 699 Aerobic methanotrophs have been classified into two phyla: the Proteobacteria which are further 700 classified into classes Alphaproteobacteria, Gammaproteobacteria, and Verrucomicrobia [98, 99]. 701 The methanotrophs in the group Gamma- and Alphaproteobacteria are also known as Type I and 702 Type II methanotrophs, respectively <sup>[97]</sup>. Type I methanotrophs can initiate CH<sub>4</sub> oxidation even 703 in lower concentration ranges (<12 ppm), hence called high-affinity methanotrophs and are more 704 commonly present in the environment. On the other hand, the Type II methanotrophs can only 705 perform CH<sub>4</sub> oxidation at higher concentrations of CH<sub>4</sub> (> 40 ppm) and hence called low-affinity 706 methanotrophs <sup>[76, 99]</sup>. Both Type I and Type II methanotrophs have been found in landfill cover 707 soils. Some commonly found Type I methanotrophic genera in landfill cover soils are 708 Methylomonas, Methylobacter, Methylomicrobium, and Methylocaldum, and Type II 709 methanotrophic genera are *Methylosinus* and *Methylocystis* <sup>[94, 100]</sup>. 710

Although, CH<sub>4</sub> oxidation in landfill cover soil is found to be a probable sink for the landfill 711 712 CH<sub>4</sub> emissions, several physical and environmental factors affect the methanotrophic activity and reduce the efficiency of the microbial CH<sub>4</sub> oxidation in landfill cover soils. Temperature, 713 714 moisture, pH, CH<sub>4</sub> availability, and aeration are some of the factors affecting CH<sub>4</sub> oxidation in 715 landfill cover soil. The optimum temperature for CH<sub>4</sub> oxidation has been found to be nearly 30 °C, however, it is found to occur even at lower temperatures (~2-6°C) showing that 716 methanotrophs can adapt to extreme temperatures <sup>[7, 93, 94]</sup>. Different studies have reported 717 different optimum temperatures for CH<sub>4</sub> oxidation however: the optimum range varies from 25-718

35 °C [101-102]. Similarly, soil moisture also affects microbial activity in the landfill cover soil as it 719 controls the diffusive ingress of gases into the soil. Scheutz and Kjeldsen<sup>[93]</sup> obtained maximum 720 CH<sub>4</sub> oxidation at moisture content range of 18–24% (w/w). At lower moisture content, CH<sub>4</sub> 721 oxidation reduces due to microbial water stress and at higher moisture contents ( $\geq 35\%$  w/w), 722 waterlogging may occur hampering the gas diffusion thereby lowering the microbial activity <sup>[93]</sup>. 723 In terms of pH, the methanotrophs perform well at the near neutral pH conditions (pH  $\sim$ 7) in 724 landfill cover soil [93, 100]. Bogner et al. [92] observed that initial CH<sub>4</sub> concentrations and oxygen 725 availability have a major effect on the oxidation rates in a landfill cover soil rather than 726 temperature and moisture. Since efficiency of CH<sub>4</sub> oxidation is affected by various physical and 727 bio-chemical factors, a need for engineering the cover systems using suitable sustainable 728 materials was soon realized, which led to extensive studies exploring alternative cover materials 729 that can enhance the CH<sub>4</sub> oxidation in landfill cover by providing favorable environmental 730 conditions for the microbial communities. 731

732

# 733 4.2.1 Cover Systems for Mitigating Methane

An international working group of scientists and researchers from Europe, USA, Canada and 734 Australia was formed in 2002 called Consortium for Landfill Emissions Abatement Research 735 (CLEAR) to address topics related to LFG generation and emissions, control and mitigation 736 strategies, prediction and modeling, microbial methane oxidation and biodegradation of other 737 NMOCs in landfill cover soils <sup>[103]</sup>. One of the focus areas of CLEAR was bio-based mitigation 738 of landfill CH<sub>4</sub> emissions. After microbial CH<sub>4</sub> oxidation in landfill cover soil was realized as a 739 potential CH<sub>4</sub> sink, suitable cover systems capable of CH<sub>4</sub> oxidation were explored as a low-cost 740 alternative to conventional covers in old landfills <sup>[104]</sup>. In the wake of enhancing microbial CH<sub>4</sub> 741

oxidation potential of landfill cover soil, the engineered bio-based systems emerged as the promising and cost-effective option for low-level CH<sub>4</sub> emissions control in landfills <sup>[76]</sup>. Initially, bio-based cover systems comprising of organic rich materials such as compost, sewage sludge, and peat were explored to create conducive environment for the methanotrophs and enhance the CH<sub>4</sub> oxidizing potential of landfill cover soil <sup>[7]</sup>. The bio-based cover systems have been explored in various forms in terms of application and operations which include biocover, biofilter and biowindow <sup>[76]</sup>.

749

#### 750 **Biofilters**

Biofilters function similar to engineered filters used for contaminants except that the landfill 751 biofilters are designed to absorb CH<sub>4</sub> by enhancing microbial CH<sub>4</sub> oxidation <sup>[76, 105]</sup>. Biofilters are 752 constructed over a certain portion of the landfill cover and require continuous feeding of LFG 753 through active or passive gas collection system <sup>[76, 106]</sup>. The biofilters are designed as fixed bed 754 reactors packed with organic media which can sustain and induce proliferation of methanotrophs. 755 Providing bio-based cover system over entire landfill may raise some issues related to 756 infiltration. Biofilters appear to be suitable option in such cases as it can be integrated with the 757 conventional cover system thereby maintaining the regulatory infiltration requirement of the 758 cover system <sup>[107]</sup>. Various biofilter media have been tested to optimize the CH<sub>4</sub> oxidation 759 efficiency of the biofilters. Gebert et al. <sup>[108]</sup> designed a biofilter integrated with landfill cover 760 761 system. The designed biofilter was an upflow system consisting of five layers (base to top): drainage gravel, expanded clay pallets, gravel, sand and topsoil (loamy sand) for vegetation 762 packed in a 15 m<sup>3</sup> polyethylene container divided into two compartments of 6 and 9 m<sup>3</sup> size. The 763 764 biofilter was connected with the passive gas vent of the landfill. CH<sub>4</sub> oxidation rates of biofilter

media were assessed through laboratory batch experiments and CH<sub>4</sub> removal rates ranged from 765 35 to 109 g CH<sub>4</sub> h<sup>-1</sup> m<sup>-3</sup>. Oxygen intrusion appeared to be an important driver for CH<sub>4</sub> oxidation 766 as it occurred at oxygen concentrations above 1.7-2.6 % ( $\nu/\nu$ ). The long-term monitoring of the 767 biofilter showed the CH<sub>4</sub> oxidation is significantly affected by the CH<sub>4</sub> influx rates with higher 768 CH<sub>4</sub> removal rates obtained for lower CH<sub>4</sub> influx <sup>[109]</sup>. Two types of biofilters: water-spreading 769 biofilter comprising of coarse sand overlain by fill sand, and vertical compost biofilter 770 comprising of a mixture of compost and polystyrene pellets were designed and evaluated for CH<sub>4</sub> 771 oxidation potential by Powelson et al. <sup>[110]</sup>. Both the biofilter designs resulted in similar CH<sub>4</sub> 772 removal efficiency (63-69%). Similarly, Abichou et al. <sup>[111]</sup> designed two types of biofilters: 773 vertical and radial, based on the direction of gas flow in the filter. The filters were housed in 774 glass barrels for protection. The filters had a drainage layer/gas distribution layer of gravel or 775 recycled glass which had the LFG inflow at the bottom. A mixture of compost and peanut foam 776 was placed over the gravel layer. The radial filter had greater surface area (459% more) than the 777 778 vertical filter which was designed to allow greater access to atmospheric oxygen and thus increase CH<sub>4</sub> oxidation. The radial biofilter resulted in higher CH<sub>4</sub> oxidation rates than the 779 vertical filter. The average percent oxidation achieved was 20% with a maximum of 100%. 780 Dever et al. <sup>[107, 112]</sup> designed a central biofiltration system with four different filters each with 781 different biofilter media and gas distribution layer. The biofilter media comprised of 1) 782 783 composted garden organics with 10% shredded wood, 2) composted MSW with 10% shredded 784 wood, 3) composted garden mix, and 4) composted MSW with 20% shredded wood. A field scale trial was set up and monitored for four years to investigate the effectiveness of the 785 biofilters. The passively aerated biofilters were able to oxidize CH<sub>4</sub> resulting in maximum and 786 787 average oxidation efficiency of 90% and 50%, respectively. CH<sub>4</sub> loading rate was found to be a controlling parameter for CH<sub>4</sub> oxidation which in turn governed the diffusive ingress of oxygen
 into the biofilter.

Some of the notable advantages of biofilters are easier implementation and requirement of 790 less maintenance during operation. Some of the challenges associated with biofilters are higher 791 CH<sub>4</sub> loading which impedes the diffusive ingress of oxygen into the filter and thus reduces the 792 793 microbial CH<sub>4</sub> oxidation potential. In the event of extreme precipitation, waterlogging may be experienced in the biofilters thereby reducing gas transport through the filter. In addition, 794 formation of exopolymeric substances (EPS) due to microbial activity may lead to clogging of 795 the biofilter media affecting the gas transport <sup>[76]</sup>. Similarly, while designing biofilters, the choice 796 of biofilter media plays an important role. For example, filter media like compost can cause 797 formation of anaerobic zones or cavities and may lead to production of CH<sub>4</sub> if moisture is not 798 regulated <sup>[109]</sup>. Therefore, materials with low organic contents make a better candidate for a 799 biofilter. 800

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#### 802 Biowindows

Biowindows are bio-based cover systems which are placed over a smaller portion of landfill 803 804 instead of the whole landfill. Biowindows are more suited at the older landfills where gas production is low, and bio-coverage is not needed over the whole expanse of the landfill <sup>[113]</sup>. 805 Unlike biofilters, biowindows do not require separate arrangement for supply of LFG as they 806 receive enough LFG directly from the waste due to the higher gas permeability and greater 807 surface area <sup>[76]</sup>. Biowindows generally have two layers: a gas distribution layer overlain by a 808 biological layer suitable for methanotrophic growth. About ten biowindows with active biologic 809 810 layer consisting of yard waste derived compost were installed at Faske landfill, Denmark and

their performance was monitored for over a year period <sup>[113]</sup>. The CH<sub>4</sub> oxidation efficiency was 811 measured before and after installation of biowindows using a mass balance approach based on 812 flux measurements as well as stable carbon isotopes method <sup>[113]</sup>. An increase in CH<sub>4</sub> oxidation 813 efficiency from 16% to 40% was observed after installation of biowindow. Similar biowindow 814 approach was followed in an older section of landfill in Austria for degasification after 815 elimination of the gas collection wells <sup>[114]</sup>. Two biowindows (8 m  $\times$  8 m each) were installed at 816 the location of previous two gas collection wells in the older section of the landfill. The 817 biowindow comprised of a lower gas distribution layer of gravel and upper biological layer 818 composed of compost mixed with wooden chips. The performance of the biowindows was 819 monitored for over 2.5 years through flux measurements and gas concentration profiling. A sharp 820 decrease in CH<sub>4</sub> emission rates were observed with the biowindow installation. 821

Although biowindow is a good alternative measure for mitigating CH<sub>4</sub> emissions from landfills, there are some challenges associated with its functional performance one of which is the lack of homogenous distribution and supply of CH<sub>4</sub> to the biowindow <sup>[113, 114]</sup>. In addition, challenges similar to biofilters such as formation of EPS and clogging of pores in the biologic layer may occur with increase in microbial activity.

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#### 828 Biocovers

Biocovers are similar to biowindows except that they are installed over the entire waste area or essentially over the entire landfill. Extensive studies have been conducted exploring various biobased materials for biocovers. Organic rich materials such as compost derived from various sources such as MSW, sewage, garden waste, as well as compost mixed with other materials such as wood chip have been explored as biocover substrates. Typically, configuration of

biocovers are close to biowindows with gas distribution layer overlain by a biologic layer and a 834 vegetative layer at the top <sup>[115]</sup>. The first biocover was tested by Huber-Humer and Lechner <sup>[104]</sup> 835 in laboratory column studies where they tested MSW compost, sewage sludge compost and 836 mixture of compost and sand. The MSW and sewage sludge compost proved to be a suitable 837 carrier for methanotrophs, however, there are certain requirements which must be met for using 838 compost in biocover, which include use of matured compost with solid organic matter and 839 minimum ammonium and salt concentrations. Ever since the testing of first biocover prototype, 840 there has been extensive number of studies exploring various organic-rich substrates for 841 biocovers. Earlier studies mainly focused on compost as the biologic layer in biocovers. Barlaz et 842 al. [82] compared the performance of biocover made of yard waste compost with soil cover in 843 landfill cells for over a period of 14 months. The static chamber technique was employed to 844 measure the fluxes at the cover site and stable carbon isotope analysis was used to evaluate the 845 in-situ CH<sub>4</sub> oxidation rate. CH<sub>4</sub> oxidation efficiencies of biocover was nearly 2.6 times more than 846 that of the soil cover. Stern et al. [116] evaluated the CH<sub>4</sub> oxidation potential of biocover 847 consisting of garden waste or pre-composted yard waste overlying a gas distribution layer of 848 crushed glass, placed over an existing 40-100 cm thick soil cover at a hotspot location with high 849 850 CH<sub>4</sub> emissions. They reported a 10-fold reduction in CH<sub>4</sub> emissions as well as two times more CH<sub>4</sub> oxidation than a control area without biocover. Biocover helped to enhance the gas retention 851 852 time as well as moisture retention in the underlying existing cover soil thereby reducing desiccation cracking and preferential flow <sup>[116]</sup>. Bogner et al. <sup>[115]</sup> analyzed the performance of a 853 biocover placed over a 15 cm thick intermediate clay cover at a landfill in Florida. Two types of 854 biocovers (deep and shallow) consisting of a 15 cm thick gas distribution layer made up of 855 856 crushed recycled glass overlain by ground garden waste (30 cm for shallow and 60 cm for deep)

were installed at the test areas. The CH<sub>4</sub> emission fluxes were monitored using static flux 857 chamber technique for over a year period. The deep biocover generally showed higher CH<sub>4</sub> 858 oxidation (ranging from 20-70%) than the shallow biocover and the soil cover. In addition, deep 859 and shallow biocovers showed higher percentage of negative fluxes showing uptake of 860 atmospheric CH<sub>4</sub> due to high CH<sub>4</sub> oxidation potential of the biocover. In the recent biocover 861 study by Lee et al. <sup>[117]</sup>, an onsite pilot biocover was set up at a sanitary landfill in South Korea 862 which consisted of a mixture of soil, perlite, earthworm cast and compost (6:2:1:1 v/v) and was 863 monitored for 240 days. The study showed the seasonal variation in temperature strongly affects 864 CH<sub>4</sub> removal efficiency with 35-43% removal efficiency in winter to 86-96% in summer. 865

Various factors affect the performance of compost as a biocover substrate. Huber-Humer 866 et al. [118] assessed the CH<sub>4</sub> oxidation potential of 30 different compost materials and their 867 mixtures and reported that bulk density, nutrient content, type of organic matter (maturity) are 868 the major factors affecting CH<sub>4</sub> oxidation rate. Similarly, Scheutz et al. <sup>[119]</sup> chose kitchen waste 869 870 derived compost over garden waste in their study due to its lower oxygen demand for their biocover study. A biocover comprising of kitchen waste derived compost was placed over an old 871 unlined landfill without gas collection system in Denmark <sup>[119]</sup>. CH<sub>4</sub> oxidation efficiency of 80% 872 873 was reported over the monitoring period of nearly two years. Discrepancies in the gas distribution in the landfill cover substantially affects the performance of the biocover leading to 874 formation of hotspots with high CH<sub>4</sub> fluxes at some portion of biocover surface <sup>[120]</sup>. In order to 875 address this issue of uneven gas distribution, Scheutz et al. <sup>[120]</sup> designed a semi-passive biocover 876 system at a landfill in Denmark, in which the compost biocover was fed by LFG collected from 877 three leachate collection wells. The biocover resulted in a CH<sub>4</sub> oxidation efficiency of 81-100%. 878 879 In addition, the authors evaluated the respiration potential of the compost and highlighted that

the surface flux may constitute CO<sub>2</sub> from respiration in addition to CH<sub>4</sub> oxidation. It suggests 880 that microbes may compete for oxygen for respiration and oxidation. One of the major 881 challenges posed by using compost in biocover is its propensity for self-degradation leading to 882 production of CH<sub>4</sub> and ultimately increasing the surface CH<sub>4</sub> flux <sup>[7]</sup>. Compost maturity was 883 identified as a major factor for the design of compost biocover [73, 104, 106] as it may lead to 884 885 increased oxygen demand and reduction in CH<sub>4</sub> oxidation if the compost used is not mature initially. Similarly, excessive formation of EPS in the zones of maximum oxidation was 886 observed by Wilshusen et al. <sup>[121, 122]</sup> leading to pore clogging and ultimately hindering oxygen 887 intrusion in the compost-based biocover. In addition, composts are rich in nutrients which may 888 not always be favorable for the methanotrophs leading to growth of other heterotrophs. In 889 addition, compost production is self-controlled as a result, it is hard to regulate their properties to 890 suit the methanotrophic growth. 891

Addressing the drawbacks of compost as a biocover substrate, an inert and recalcitrant 892 substrate called biochar was explored for landfill CH<sub>4</sub> mitigation. Biochar is a solid product 893 produced by pyrolysis of waste biomass in the oxygen deficient condition at temperatures 894 ranging from 300-1000 °C<sup>[123, 124]</sup>. The properties of biochar vary depending on the conditions of 895 pyrolysis and types of feedstock <sup>[125, 126]</sup>. A wide variety of feedstocks can be used for biochar 896 production such as wood chips, manure, and MSW. Biochar is characterized with high surface 897 area and porosity, and adsorption potential for various chemical compounds including organic 898 pollutants and gases <sup>[95, 127]</sup>. Owing to its unique physicochemical properties, use of biochar has 899 gained prominence in various environmental applications <sup>[123]</sup>. The use of biochar in soil drew 900 901 attention for carbon sequestration and mitigating climate change by replacing the organic

biomass with less degradable form of carbon <sup>[126, 128]</sup>. Biochar had long been used in agriculture
to improve the physical properties of the soil, fertility, and crop yield <sup>[128, 129]</sup>.

Biochar as a biologic amendment to landfill cover soil to mitigate fugitive CH<sub>4</sub> emissions 904 was first explored by Yaghoubi et al. <sup>[127]</sup> and Reddy et al. <sup>[95]</sup>. Laboratory column experiments 905 were performed to simulate biocover with biochar-amended soil. The simulated biocover 906 907 comprised of gas distribution layer (gravel) overlain by pinewood-based biochar-amended soil (20/80 weight%) and exposed to synthetic LFG for 4 months with variable flux rates <sup>[95]</sup>. High 908 CH<sub>4</sub> oxidation rates were attained in biochar-amended soil layers with higher abundance of 909 methanotrophic communities. The maximum CH<sub>4</sub> oxidation was observed at the upper 0-30 cm 910 of the biochar-amended soil cover. Since, these studies established that biochar can enhance the 911 CH<sub>4</sub> oxidation potential of the landfill cover soil by increasing oxygen availability and providing 912 habitable environment for the methanotrophs, further investigations were carried out to establish 913 the optimum biochar amendment ratios to economize the biocover application at landfills. In this 914 regard, Yargicoglu and Reddy<sup>[130, 131]</sup> performed a series of large-scale column experiments with 915 variable biochar amendment ratios and different cover configurations and assessed the long-term 916 performance of the biochar-amended soil under dynamic gas flow conditions. Yargicoglu and 917 Reddy <sup>[131]</sup> tested 2% and 10% biochar amendment to landfill cover soil and varied the biochar 918 amendment depth. In two of the columns, biochar amendment (2% and 10%) were applied at 20-919 920 40 cm depth from ground surface of 60 cm thick biocover. One column had 10% biocharamended soil over entire 60 cm of biocover thickness and another column had 60 cm of soil 921 (control). Batch incubations were performed on the samples exhumed from various depths of 922 each of the two columns to evaluate the CH<sub>4</sub> oxidation potential of the biochar-amended soil 923 924 samples. The 10% biochar-amended soil showed the highest CH<sub>4</sub> removal potential for wide

range of CH<sub>4</sub> loads (50- 200 g CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) during the incubation period of 478 days. Biochar-925 amended soil showed characteristically higher moisture retention capacity reducing desiccation 926 cracking which was otherwise very prominent in soil control column. Maximum CH<sub>4</sub> oxidation 927 rate of 270 µg CH<sub>4</sub> g<sup>-1</sup>d<sup>-1</sup> was observed in 10% biochar-amended soil cover at 30 cm depth. 928 Yargicoglu and Reddy <sup>[130]</sup> also evaluated the effect of providing biochar alone as a thin layer 929 within the soil cover. They observed that biochar helps to increase the moisture retention and 930 931 provides favorable conditions for methanotrophs to proliferate, but biochar, being an inert material, does not increase CH<sub>4</sub> oxidation if used alone in the cover. Therefore, for an enhanced 932 933 CH<sub>4</sub> oxidation, biochar needs to be mixed with landfill cover soil. In addition to laboratory column tests, field-scale pilot tests were performed to evaluate the performance of biochar-based 934 biocover <sup>[99, 132]</sup> under real landfill conditions. Eight test plots including four replicate plots with 935 60 cm thick biocover and 30 cm thick gravel gas distribution layer placed over the existing 30 936 cm thick intermediate cover soil layer were tested over a span of 8 months (September to May) 937 at a landfill in Illinois. Biocover profiles with 2% and 10% (w/w) biochar-amended soil were 938 tested in the pilot-scale field tests. Surface emissions were analyzed with static flux chamber and 939 the gas concentration along the depth of the cover through gas probes clusters. The field tests 940 were highly impacted by the heterogeneity of the waste which led to very low CH<sub>4</sub> loads at the 941 biochar-based biocover test plots and high CH<sub>4</sub> loads at the soil control test plots. Due to the high 942 943 differences in the CH<sub>4</sub> loading from the waste, soil core samples showed high CH<sub>4</sub> oxidation rates in batch incubation than the biochar-amended soil samples <sup>[99]</sup>. This observation was further 944 supported by the high relative abundance of the methanotrophic genera in the samples from soil 945 control test plot. Type I methanotrophs were more abundant than Type II methanotrophs in the 946 field samples as well as the laboratory column samples [99]. The effect of irregular gas 947

distribution on biocover performance had previously been identified in several studies such as Scheutz et al. <sup>[120]</sup> who designed a semi-passive biocover system to deal with the uneven gas distribution from the waste. High variability in CH<sub>4</sub> oxidation rates and diversity in methanotrophic communities due to heterogeneity of waste at landfills had been reported in prior field studies <sup>[133, 134]</sup>.

In a recent study by Huang et al. <sup>[135]</sup>, biochar derived from cottonwood shavings were 953 used for amending landfill cover soil for CH<sub>4</sub> oxidation. Column experiments (along with soil 954 control) were performed by amending cover soil with 15% ( $\nu/\nu$ ) biochar at 10-30 cm depth and 955 performance was monitored for over a period of 101 days. Methanotrophic culture was poured at 956 every 5 cm depth of soil to enrich the soil with MOB prior to exposure to synthetic LFG. In 957 addition, one of the biocover test columns was supplied with additional aeration from a point 958 below the biochar- amended soil layer besides air supply from the top of the column to increase 959 the oxygen supply. Biochar-amended soil columns showed higher CH<sub>4</sub> oxidation rates (90.6% -960 85.2%) than the soil control (78.6%). The authors observed a reduction in the CH<sub>4</sub> oxidation 961 rates at the later stage of the experiment which was attributed to the depletion of the nitrogen 962 nutrient in the soil. The relative abundance of Type II methanotrophs was more than that of Type 963 I which contrasts with the observations of Reddy et al. <sup>[99]</sup> where Type I methanotrophs were 964 reported to be more abundant, and the reason could be likelihood of a shift in microbial 965 community due to the formation of microaerophilic environment from EPS formation which 966 favors the growth of Type II methanotrophs <sup>[122]</sup>. Another recent study explored hydrophobic 967 biochar-amended soil through laboratory column tests and compared its CH<sub>4</sub> oxidation potential 968 with hydrophilic biochar amended soil and soil control <sup>[136]</sup>. The rice straw-based biochar was 969 970 pretreated to render hydrophobic properties to tackle the high hydraulic conductivity of hydrophilic biochar. The authors observed high CH<sub>4</sub> oxidation rates in biochar-amended soil
than the soil control with hydrophobic biochar attaining similar oxidation rates as hydrophilic
biochar.

Most of the biochar-based biocover studies are based on the laboratory column studies 974 which represent more idealized conditions whereas in the field the gas flow conditions as well as 975 976 meteorological conditions vary severely which ultimately affect the performance of the biocover system. There have not been many field-scale studies performed till date with biochar-amended 977 soil cover which have limited its full-scale application for a landfill closure. Although, biochar-978 amended soil cover appears to be effective for enhancing landfill CH<sub>4</sub> oxidation and mitigating 979 CH<sub>4</sub> emissions, there is a need for extensive field studies to establish the performance of biochar-980 amended soil under various climatic conditions. 981

One of the issues that could impact the performance of the biocovers and 982 biowindows could be the contrasting difference in the permeabilities of CH<sub>4</sub> oxidizing layer and 983 gas distribution layer giving rise to capillary effect. Many studies<sup>[137, 138]</sup> have reported formation 984 of capillary barrier and accumulation of water at the interface of CH<sub>4</sub> oxidizing layer and gas 985 distribution layer resulting in uneven gas distribution and formation of CH<sub>4</sub> hotspots. Although 986 987 capillary effect may be inexorable due to the functional requirements of biocover, it can be minimized by proper selection of the materials and designing the biocovers to minimize clogging 988 due to water at the interfaces. For example, Cassini et al.<sup>[138]</sup> designed the gas distribution layer 989 990 in zig-zag shape to allow water to collect at low points and break the capillary barrier due to pressure build up. 991

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#### 993 4.2.2 Cover Systems for Mitigating Non-Methane LFG Components

Apart from CH<sub>4</sub>, LFG comprise of other trace organic compounds such as alkanes, alkenes, 994 aromatic hydrocarbons, halogenated aliphatic compounds, and organic sulfur compounds <sup>[82, 139]</sup> 995 which pose serious threats to environment as well as human health. The volatile organic 996 compounds (VOCs) generated in the landfill are the outcome of the anaerobic decomposition of 997 the organic waste in the limited oxygen environment <sup>[140]</sup>. The CH<sub>4</sub> oxidation potential of landfill 998 cover soil suggested that some organic compounds may also be oxidized under similar 999 conditions which prompted many researchers to explore biocovers for mitigation of other 1000 1001 NMOCs in LFG. Table 1 summarizes some of the studies which investigated the attenuation of NMOCs present in the LFG through biologic degradation. Some of the aromatic hydrocarbons 1002 such as benzene and toluene are rapidly oxidized in aerobic conditions and they show maximum 1003 removal in the oxic regions of the soil cover <sup>[143]</sup>. Methanotrophs can co-metabolize various 1004 hydrocarbons including halogenated aliphatic hydrocarbons <sup>[139]</sup>. Scheutz and Kjeldsen <sup>[142]</sup> 1005 investigated of 1006 the degradation potential chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) along with CH4 in laboratory column experiments and soil 1007 microcosms in landfill cover soil. High degradation potential of both HCFCs and CFCs was 1008 1009 observed in column experiments with HCFCs degraded in aerobic conditions and CFCs in anaerobic conditions. In another study by Scheutz et al. <sup>[139]</sup>, an inverse relation between 1010 1011 chlorine/carbon ratio and degradation rates was observed. The lower chlorinated carbons such as 1012 vinyl chloride (VC) had highest degradation rates and trichloroethylene (TCE) had the lowest with no degradation of polychlorinated ethylene (PCE). Although fully substituted aliphatic 1013 1014 compounds such as PCE and CFC-11 show limited oxidation in aerobic conditions, they form 1015 intermediate compounds like VC under anaerobic condition which are more prone to aerobic

degradation <sup>[89]</sup>. Since methanotrophic conditions are suitable for degradation of several NMOCs, the use of biocovers (such as compost) have shown to favor the reduction of NMOC emissions in landfills over the soil covers <sup>[82]</sup>. Wang et al. <sup>[144]</sup> also observed a positive correlation between CH<sub>4</sub> oxidation and NMOCs attenuation. Although, laboratory-scale studies have shown potential for degradation of various NMOCs in landfill cover soils and biocovers, the same needs to be verified with extensive field demonstrations.

In addition to GHG emissions and NMOCs, odor is another challenging concern of the 1022 landfills. Odor problems impose environmental as well as societal impacts such as degradation of 1023 1024 quality of life, depreciation of the property value, health risks to workers as well as people living near the landfills <sup>[146]</sup>. The odorous components of LFG comprise of phenols, nitrogen 1025 compounds, sulfur compounds, VOCs, and organic acids <sup>[117, 147]</sup>. Of the many odorous 1026 compounds, H<sub>2</sub>S, methyl mercaptan, dimethyl sulfide and dimethyl disulfide are considered the 1027 major odor causing compounds <sup>[148, 149]</sup>. Among the sulfur compounds, H<sub>2</sub>S has the highest 1028 abundance (~80%) and thus, a major contributor of the odor around landfills <sup>[146, 149]</sup>. In addition, 1029 H<sub>2</sub>S is also considered the most perceptible odorous compound with the unique stench of rotten 1030 egg<sup>[150]</sup>. 1031

Non methane LFG **Observations** Type of cover Type of study Reference material components Barlaz et al. [82] Selected NMOCs and Soil cover (clay) and Field Most of the highest emissions of NMOCs and speciated organic speciated organic biocover (yard waste investigations compounds measured were from soil compounds compost) (halogenated aliphatics, cover. The suitable conditions induced aromatics, halocarbons) by biocover promotes aerobic degradation of organic compounds other than methane. Scheutz et al. [89] Selected VOCs Soil cover (loamy Laboratory Lower chlorinated compounds are (chlorinated alkanes, batch more prone to aerobic degradation. sand) incubations alkenes, halocarbons, Oxidation rates of aromatics) Halogenated aliphatics ranged from 0.03-1.7 µg g soil<sup>-1</sup> h<sup>-1</sup> • Aromatics ranged from 0.17-1.4 µg  $g_{soil}^{-1} h^{-1}$ ) The compost derived simulated covers Scheutz et al. <sup>[106]</sup> Selected VOCs (CFC-Compost derived Laboratory 11, HCFC-21, HCFCshowed significant attenuation capacity batch cover for the VOCs (56-94 % removal 31, and (compost/wood incubation and fluoromethane (HFCchips (1:1), dynamic efficiency in compost/woodchip). compost/sand (1:1), 41)) column compost/sand(1:5),experiments and supermuld) Soil cover (Coarse Scheutz et al. [139, 141] Selected NMOCs Laboratory Oxidation rates for (halogenated aliphatics, sand + silty to sandy halogenated aliphatic compounds batch aromatics) loam) incubation ranged from 0.06 to 8.56  $\mu$ g g soil<sup>-1</sup>  $d^{-1}$ tests and Field benzene and toluene were 28 and

1033 **Table 1.** Summary of studies exploring alternative covers for mitigating non methane compounds from landfill gas

#### 1032

		investigations	39 $\mu$ g g soil <sup>-1</sup> d <sup>-1</sup> , respectively	
Halocarbons (CFC-11, CFC-12, HCFC-21, and HCFC-22)	Soil cover (Loamy sand to sandy loam)	Laboratory column experiments and batch incubation tests	<ul> <li>Anaerobic degradation of CFCs (90% and 30% removal of CFC-11 and CFC-12, respectively) with no degradation under aerobic conditions</li> <li>Aerobic degradation of HCFCs (61% and 41% removal of HCFC- 21 and HCFC-22, respectively)</li> </ul>	Scheutz and Kjeldsen <sup>[142]</sup>
Selected VOCs (TeCM, TCM, DCM, TCE, VC, benzene, toluene)	Soil cover (loamy sand, sandy loam)	Laboratory batch incubations and column experiments	Degradation of all chlorinated compounds with removal efficiency > 57%, high degradation rates of benzene and toluene (0.18 and 0.12 g m <sup>-2</sup> d <sup>-1</sup> , respectively)	Scheutz and Kjeldsen <sup>[143]</sup>
Selected NMOCs (sulfur compounds, halogenated hydrocarbons, aromatics, aliphatic hydrocarbons, terpene, oxygenated compounds)	Intermediate cover soil	Field investigations	Significant reduction in concentrations of NMOCs (halogenated compounds and aromatics) observed in the soil cover. A synergistic effect of methane oxidation on NMOCs degradation was observed	Wang et al. <sup>[144]</sup>
Selected NMVOCs (toluene)	Waste biocover soil and landfill cover soil (sandy loam)	Laboratory column experiments	Waste biocover soil (99.7-99.9%) showed relatively higher toluene removal potential than landfill cover soil (97.8-99.6%). Toluene removal was higher in the absence of methane due to lesser competition for available oxygen.	Su et al. <sup>[145]</sup>

Significantly higher  $H_2S$  concentrations are prevalent in the construction and demolition 1034 (C&D) landfills due to the biological degradation of gypsum (CaSO<sub>4</sub>. 2H<sub>2</sub>O), a prime component 1035 of C&D waste, by the sulfate-reducing bacteria (SRB) <sup>[151]</sup>. Since C&D waste landfills do not 1036 have strict gas collection regulations as MSW landfills, the odorous gases are more likely to 1037 escape to the atmosphere <sup>[152]</sup>. H<sub>2</sub>S is also generated in notable amounts in MSW landfills from 1038 sulfur containing organic waste such as paper and food, and sludge from wastewater treatment 1039 plants <sup>[153]</sup>. Although H<sub>2</sub>S is produced in trace amounts in MSW landfills, H<sub>2</sub>S can be perceived 1040 at very low concentrations (odor threshold of 0.01-1.5 ppmv) and can be harmful at low 1041 concentrations of 30-40 ppmv<sup>[154]</sup>. The concentrations can reach 450 ppmv and more in landfill 1042 cells <sup>[152]</sup>. 1043

Although alternative landfill cover systems with wide variety of substrates were 1044 developed, most of them focused on biological CH<sub>4</sub> oxidation. As the odor nuisance from 1045 landfills started to emerge as a pressing challenge, alternative cover systems were explored to 1046 mitigate odor and reduce releases of odorous compounds. Plaza et al. <sup>[152]</sup> performed laboratory-1047 scale column experiments to simulate the H<sub>2</sub>S production from C&D waste and tested five 1048 different cover materials: 1) sandy soil, 2) lime amended sandy soil, 3) clayey soil, 4) fine 1049 1050 concrete (particle size < 2.5 mm), and 5) coarse concrete (particle size > 2.5 cm) by placing the 1051 cover materials over the waste in the experimental columns.  $H_2S$  production from the waste 1052 ranged from 5% to 15% ( $\nu/\nu$ ). Lime amended sandy soil and fine concrete showed highest H<sub>2</sub>S removal efficiencies (> 99%) followed by clayey (65%) and sandy soils (30%) with coarse 1053 concrete showing lowest H<sub>2</sub>S removal efficiency. The reduction in H<sub>2</sub>S emission through clayey 1054 1055 and sandy soil cover was attributed to the formation of physical containment by the cover system 1056 limiting the diffusive migration of  $H_2S$ .  $H_2S$  removal in lime amended soil and fine concrete was

1057 attributed to the reaction between hydrated lime and  $H_2S$  forming sulfide minerals (**Eqs. 3** and **4**) 1058 under alkaline conditions induced by the lime and concrete <sup>[152]</sup>. Although mineralogical 1059 identification was not done to assert the formation of sulfides, the change in color of the cover 1060 substrates to black towards the end of the experiment was taken as an indication of formation of 1061 the metal sulfides.

- 1062  $Ca(OH)_2 + H_2S \rightarrow CaS + 2H_2O$  (3)
- 1063  $H_2S + CaO \rightarrow CaS + H_2O$  (4)

Similarly, Xu et al. <sup>[151]</sup> performed field tests as well as laboratory microcosm tests to evaluate 1064 the attenuation of  $H_2S$  by six different alternative cover materials: 1) sandy soil, 2) fine concrete, 1065 3) compost, 4) sandy soil amended with 10% agricultural lime (CaCO<sub>3</sub>), 5) sandy soil amended 1066 1067 with 1% hydrated lime  $[Ca(OH)_2]$  and 6) sandy soil amended with 3% hydrated lime. All the covers were able to remove H<sub>2</sub>S (99%) during the field-testing period of ten months, however, 1068 sandy soil cover showed the lowest removal rates among the six cover systems. In the laboratory 1069 1070 batch experiments, fine concrete showed a rapid H<sub>2</sub>S removal rate (90% within 5 mins of exposure). Similarly, the hydrated lime (3% and 1%) amended soil took 10 mins for 90% 1071 removal of H<sub>2</sub>S. Sandy soil showed the lowest H<sub>2</sub>S removal capacity, 60% in 60 mins. The 1072 removal of H<sub>2</sub>S by concrete and lime-amended soils was attributed to the adsorption on the 1073 particle surface and conversion to metal sulfides as reported by Plaza et al. <sup>[152]</sup>. Compost showed 1074 substantial H<sub>2</sub>S removal both in field and laboratory tests. Although it did not have high 1075 alkalinity as that of lime, the H<sub>2</sub>S removal was attributed to the biotransformation by sulfur 1076 oxidizing bacteria (SOB) which was supported by reduction in pH from 7.4 to 6.3<sup>[151]</sup>. 1077

1078**Table 2** summarizes some of the studies which studied potential of various alternative1079cover materials for mitigating odorous compounds including  $H_2S$  in LFG. A wide range of

biological materials such as waste biocover soil, compost, mixtures of soil and earthworm casts 1080 have shown appreciable adsorption capacity for malodorous compounds such as H<sub>2</sub>S, 1081 trimethylamine, aldehydes, etc. (Table 2). The prime mechanism for H<sub>2</sub>S removal in biocovers 1082 and landfill cover soils has been attributed to adsorption and biotransformation <sup>[149, 151, 155]</sup>. Xia et 1083 al. <sup>[149]</sup> observed high diversity of SOB such as *Halothiobacillus*, *Thiobacillus*, *Thiovirga* and 1084 Bradyrhizobium and SRB such as Desulfobacca, Desulforhabdus and Syntrophobacter in the 1085 landfill cover soils. SOB oxidizes  $H_2S$  to elemental sulfur (S<sup>0</sup>) and sulfate (SO<sub>4</sub><sup>2-</sup>) under aerobic 1086 conditions <sup>[158]</sup>. SRBs are mostly anaerobes but they may be present in oxic environments as 1087 some of the species show oxygen tolerance <sup>[159]</sup>. Xia et al. <sup>[158]</sup> studied the sulfur metabolizing 1088 bacteria in the waste biocover soil and landfill cover soil and observed 4.3 to 5.4 times increase 1089 in sulfur oxidation rate in waste biocover soil in comparison to landfill cover soil. pH affects the 1090 abundance of SOB and SRR in the soil and a neutral or slightly alkaline pH has been found to be 1091 favorable for oxidation of sulfur in landfill cover soils <sup>[149]</sup>. Many studies have shown the 1092 potential in biocovers and landfill cover soils for mitigation of odorous compounds emanating 1093 from landfills, however there is a need for exploring an integrated system which can mitigate all 1094 the major LFG components such  $CH_4$ ,  $CO_2$  and  $H_2S$  at once as it is not feasible and economical 1095 1096 to place separate cover systems in a landfill to mitigate each LFG component.

**Target Compound Cover Material Key Observations** Reference Lee et al. [117] Ammonia, hydrogen Biocover made of • The removal efficiency of odorous compounds was sulfide, methyl mixture of soil, perlite, higher in biocover and achieved nearly 85% in all mercaptane, acetic earthworm castings, seasons. aldehyde, toluene, compost, 6:2:1:1, v/v) • Among the 22 odorous compounds tested, biocover xylene and other 16 showed highest removal efficiency for H<sub>2</sub>S across alls odorous compounds the seasons. Mechanism for H<sub>2</sub>S removal attributed to adsorption and biodegradation. Sulfur oxidizing bacteria (SOB) such as Arthrobacter • were identified in the biocover which can oxidize sulfur compounds like H<sub>2</sub>S, dimethylsulfide and dimethyldisulfide to sulfate. Total reduced sulfur Four cover types tested: The odor and TRS removal efficiency were > 95% in Capanema et al. [146] 1) mixture of sand-(TRS) compounds all four cover systems. compost and gravel; 2) The major parameters affecting the removal efficiencies Mixture of sandwere degree of saturation and gas loading rate. The compost; 3) sand odor concentrations following significant precipitation overlain by topsoil; and events (48- hour accumulated precipitation) were 4) mixture of topsoil and lower. compost underlain by • Higher biogas loading resulted in increased emissions topsoil and sand layers of odor as well as TRS compounds. He et al. [154] Waste biocover soil, Hydrogen sulfide • Highest adsorption capacity shown by waste biocover landfill cover soil. soil in comparison to landfill cover soil, mulberry soil, mulberry soil and sandy and sandy soil. soil Maximum adsorption by waste biocover under • optimum temperature, moisture and pH condition was

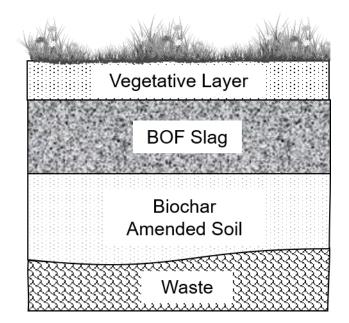
**Table 2.** Summary of studies exploring alternative covers for mitigation of malodorous compounds from landfill gas

		$60 \pm 1 \text{ mg/kg}.$	
Hydrogen sulfide	Waste biocover soil and landfill cover soil	<ul> <li>H<sub>2</sub>S removal efficiency of greater than 90% achieved in both waste biocover soil and landfill cover soil, however waste biocover soil showed better removal efficiency among the two cover systems.</li> <li>The major H<sub>2</sub>S removal mechanism was adsorption of H<sub>2</sub>S gas on particle surface, dissolution into pore water and biotransformation.</li> <li>Sulfide was observed as the major H<sub>2</sub>S removal.</li> <li>The growth of sulfur oxidizing bacteria and sulphate reducing bacteria were observed with the exposure of the covers to H<sub>2</sub>S.</li> </ul>	He et al. <sup>[155]</sup>
Trimethylamine (TMA), and dimethyl sulfide (DMS)	Biocover made of mixture of tobermolite, landfill cover soil, and earthworm castings (2:1:1, w/w)	<ul> <li>The malodorous components were completely removed by the cover system with the removal efficiencies of 100%.</li> <li>The major mechanism was oxidation and the removal of the components started from the bottom of the cover system (40-50 cm below top surface).</li> </ul>	Lee et al. <sup>[156]</sup>
Hydrogen sulfide	Four different cover materials tested: 1) Charcoal sludge compost (CSC); 2) final landfill cover soil (FCS); 3) Aged refuse (AR); and 4) Clay soil (CS)	<ul> <li>CSC showed highest H<sub>2</sub>S removal in both laboratory (~88%) and field studies (~82%) followed by FCS (81- 68%) and AR (77-59%) with CS showing lowest removal efficiency (72-50%).</li> <li>The H<sub>2</sub>S removal was attributed to adsorption, chemical reactions, and biological oxidation.</li> <li>Significant increase in sulfate and total sulfur concentrations observed in CSC confirming biotransformation of H2S by Sulfur oxidizing bacteria.</li> </ul>	Ding et al. <sup>[157]</sup>

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## 1100 4.2.3 Biogeochemical cover system

Most of the alternative cover systems have been studied with respect to mitigation of CH<sub>4</sub> or 1101 NMOCs or odor. Although, CO<sub>2</sub> constitutes 50% of the volume of LFG and is a highly potent 1102 GHG, very little focus is given to landfill CO<sub>2</sub> mitigation. In addition, mitigating CH<sub>4</sub> alone or 1103 1104 odor alone does not solve the problems of fugitive landfill emissions. In this regard, an alternative cover system called biogeochemical cover is being developed which comprise of 1105 biochar-amended soil and basic oxygen furnace (BOF) steel slag <sup>[160-162]</sup>. The biogeochemical 1106 cover system leverages on the CH<sub>4</sub> oxidation potential of biochar-amended soil <sup>[131]</sup> and the CO<sub>2</sub> 1107 and H<sub>2</sub>S sequestration potential of BOF slag <sup>[162, 163-166]</sup> to mitigate CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>S 1108 simultaneously thus rendering MSW landfills nearly emissions free. A schematic of the 1109 biogeochemical cover system is shown in Fig. 8. CH<sub>4</sub> is oxidized into CO<sub>2</sub> in the biochar-1110 amended soil layer of the biogeochemical cover. Biochar amendment assists in enhancing the 1111 CH<sub>4</sub> oxidation potential of the soil as discussed in the biocover section earlier. The CO<sub>2</sub> 1112 produced during oxidation of CH<sub>4</sub> in the biochar-amended soil layer and CO<sub>2</sub> and H<sub>2</sub>S generated 1113 in the waste passes through the overlying BOF slag layer where it gets sequestered by the 1114 1115 geochemical reaction mechanisms. BOF slag is a type of steel making slag which is generated as the byproduct during steel making process. The similarity of steel slag's chemical composition to 1116 1117 natural minerals capable of binding CO<sub>2</sub> naturally led to intensive studies exploring CO<sub>2</sub> sequestration potential of steel slag <sup>[167]</sup>. The peculiar properties of BOF slag such as high 1118 alkalinity (pH > 11), presence of Ca, Mg and iron (Fe) containing minerals, and high shear 1119 1120 strength properties make it suitable for use in landfill cover applications. The Ca containing 1121 minerals such as lime (CaO), portlandite [Ca(OH)<sub>2</sub>] and larnite (Ca<sub>2</sub>SiO<sub>4</sub>) present in the BOF



1123

1124 Figure 8. Schematic of biogeochemical cover system

1125

slag can readily bind CO<sub>2</sub> and convert it into stable form of carbonates (CaCO<sub>3</sub>) as shown in the

- 1127 Eqs. 5–7.
- 1128  $CaO + CO_2 \rightarrow CaCO_3$  (5)
- 1129  $Ca(OH)_2 + CO_2 \rightarrow CaCO_3$  (6)

1130 
$$Ca_2SiO_4 + 2CO_2 \rightarrow 2CaCO_3 + SiO_2 (7)$$

1131 Reddy et al. <sup>[163-165]</sup> investigated CO<sub>2</sub> sequestration potential of the BOF slag under various 1132 conditions such as moisture, temperature, LFG loading rates, LFG conditions (dry and humid), 1133 BOF slag types, and slag particle sizes (fine to coarse). BOF slag showed significant potential for 1134 CO<sub>2</sub> sequestration under simulated LFG conditions. Moisture appeared to be an important 1135 parameter for initiation of the carbonation reactions in the slag however, the moisture content as 1136 low as 10% (*w/w*) resulted in significant carbonation (68 g CO<sub>2</sub>/kg BOF slag) in the laboratory

batch experiments <sup>[162]</sup>. A prolonged carbonation was observed upon exposure to continuous 1137 flow of humid LFG (mixture of 50% CH<sub>4</sub> and 50% CO<sub>2</sub>, v/v) resulting in CO<sub>2</sub> sequestration of 1138 350 g CO<sub>2</sub>/kg BOF slag <sup>[163]</sup>. Apart from moisture, particle size and BOF slag type have 1139 pronounced effect on the CO<sub>2</sub> sequestration capacity of the BOF slag. In the study by Reddy et 1140 al. <sup>[165]</sup>, fine slag (mean particle size = 0.094 mm) showed highest CO<sub>2</sub> sequestration (255 g 1141 1142  $CO_2$ /kg BOF slag) followed by the slag with original gradation as obtained from the steel plant (mean particle size = 0.47 mm) which was 155 g CO<sub>2</sub>/kg BOF slag. Coarse slag (mean particle 1143 size = 3.05 mm) showed the lowest CO<sub>2</sub> sequestration potential (66 g CO<sub>2</sub>/kg BOF slag). The 1144 properties of BOF slag vary depending on the production batch and the proportions of the 1145 fluxing agents charged during the steel making process <sup>[164, 168]</sup>. In the study by Reddy et al. <sup>[164]</sup>, 1146 the BOF slags obtained from different plants showed different CO<sub>2</sub> sequestration potential which 1147 was mainly attributed to the heterogeneity in the mineralogical composition and the average 1148 particle sizes of the BOF slag. Overall, BOF slag appeared to be a promising alternative for CO<sub>2</sub> 1149 sequestration from LFG. 1150

1151 Several studies have explored  $H_2S$  removal potential of steel slag <sup>[166, 169-173]</sup>. All the 1152 studies showed a promising potential for  $H_2S$  removal in steel slag. Iron appeared to be the 1153 leading metal in binding  $H_2S$  in the form of iron sulfides (**Eqs. 8–9**).

1154  $FeO + H_2S \leftrightarrow FeS + H_2O$  (8)

1155 
$$Fe_2O_3 + 3H_2S \to Fe_2S_3 + 3H_2O$$
 (9)

Since steel slag has abundant iron content in the form of iron oxides, it makes a suitable alternative for mitigating H<sub>2</sub>S at the landfills. Recently, Chetri et al. <sup>[166]</sup> performed a series of laboratory batch and column experiments with BOF slag under various simulated LFG conditions (48.25% CH<sub>4</sub>, 50% CO<sub>2</sub> and 1.75% H<sub>2</sub>S  $\nu/\nu$ ). The BOF slag was able to sequester both CO<sub>2</sub> and H<sub>2</sub>S resulting in maximum CO<sub>2</sub> and H<sub>2</sub>S removal potential of 300 g CO<sub>2</sub>/ kg BOF slag and 38 g H<sub>2</sub>S/kg BOF slag, respectively.

The highly alkaline nature of steel slag may be favorable for CO<sub>2</sub> and H<sub>2</sub>S sequestration, but 1162 it may also impede the survival of methanotrophic community for CH<sub>4</sub> oxidation in the 1163 1164 biogeochemical cover, as the optimum pH for CH<sub>4</sub> oxidation has been reported to be in the range of 6.5-7.5 <sup>[93, 100]</sup>. Although, the biogeochemical cover proposed by Reddy et al. <sup>[160, 161]</sup> aims to 1165 have biochar-amended soil and steel slag layers in separate layers, there is still concern for effect 1166 1167 of infiltrated water percolating through the slag layer on the microbial CH<sub>4</sub> oxidation in underlying biochar-amended soil. Hence, Reddy et al. [174] investigated the effect of slag 1168 infiltrated water on CH<sub>4</sub> oxidation and microbial community in the landfill cover soil. The results 1169 from the study showed that the slag infiltrated water did not have a significant impact on the CH<sub>4</sub> 1170 oxidation potential due to the high buffering capacity of the landfill cover soil. Moreover, the 1171 1172 slag infiltrated water did not impact the microbial community composition substantially even at the highest concentration of the slag infiltrated water (100% of the soil's moisture content of 1173 20% w/w). Therefore, using BOF steel slag as a CO<sub>2</sub> and H<sub>2</sub>S sequestering layer in the 1174 1175 biogeochemical cover does not appear to have a negative impact on the microbial CH<sub>4</sub> oxidation of the biochar amended soil layer. However, the effect of BOF slag carbonation on the cover's 1176 1177 porosity, hydraulic conductivity, volumetric stability, etc. during long-term operation needs to be 1178 evaluated as these may significantly affect the performance of the cover system. Studies have reported reduction in the porosity of the BOF slag due to calcite precipitation <sup>[175, 176]</sup>. Although 1179 1180 reduction in pore size may suggest that the carbonation may result in reduction of hydraulic 1181 conductivity of the carbonated slag, the studies confirming the same are scarce. On the other

hand, studies<sup>[177]</sup> have also reported increase in particle size after carbonation due to particle aggregation and volume expansion increasing average pore size and gas diffusion. Hence, there is a paucity of information asserting the behavior of BOF slag under actual landfill condition. Therefore, there is a need to evaluate the engineering behavior of carbonated slag for a sustainable cover design. Similarly, the performance of biogeochemical cover needs to be validated with the help of field investigations under dynamic meteorological field conditions.

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### 1189 **5 Research Challenges**

Over the years, the landfilling practices have evolved with significant technological advances in landfill cover systems. Modern engineered cover systems with very low permeability and high strength have been developed with the advanced geosynthetic designs and performance. Parallelly, there have been remarkable advancements in the alternative cover systems. However, the literature unveils some key challenges associated with the modern engineered cover systems and the alternative cover systems. Some of the research challenges are described as follows:

Resiliency: With the extreme climatic events becoming recurrent and landfill cover being directly exposed to the environmental conditions, there is a need to develop cover
 systems which can perform their design functions and meet regulatory requirements even under the extreme climatic events such as flooding, draught, excessive snow, etc.

Elevated temperatures: The issues of elevated temperature landfills where the
 temperatures of waste within the landfill exceeds well over 65 °C <sup>[178, 179]</sup> are gaining
 wide prominence. Hence, the performance of conventional and bio-based cover systems
 under elevated temperature conditions needs investigation.

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• Emerging pollutants: As the waste composition is changing continually, more and more emerging contaminants are being recognized. Hence, the future research needs to focus on developing cover systems to mitigate these emerging pollutants from LFG emissions.

 Economic feasibility: Landfilling practices vary significantly in low income countries or developing countries where open dumping forms a popular waste disposal alternative with nearly 93% of wastes being openly dumped <sup>[1]</sup>. Similarly, the type of waste varies geographically and thus the gas composition and gas emission rates. Hence, the landfills designed for one geographic region or income region might not be attainable for another region. It is a challenge for researchers to develop cover systems which can serve in wide range of economic and geographic conditions.

Management of abandoned landfills: The modern engineered landfills are designed to 1214 • comply with the stringent regulatory requirements however, the abandoned landfills 1215 1216 which do not have necessary components such as an impermeable bottom liner system, leachate collection system and impermeable cover system to meet the regulatory 1217 requirements are often the cause for concern. Although the majority of LFG, mainly CH<sub>4</sub>, 1218 is generated during first few years of waste disposal, the gas generation continues for 1219 several hundred years after the closure of the landfill <sup>[180]</sup>. Hence, landfills without proper 1220 enclosure systems in place such as abandoned landfills may pose threat to human health 1221 and environment for a prolonged period. Therefore, it presents a challenge for current and 1222 future researcher to develop cover systems which are not only deemed serviceable for 1223 1224 modern engineered landfills but also to the abandoned landfills without imposing significant economic burden on the managers of abandoned landfills. 1225

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• Longevity and durability: Because landfills emit gases for a prolonged duration of time 1226 after closure, it is utmost important that the alternative covers perform their function for 1227 the entire service life of the landfill. For example, some bio-based cover materials such as 1228 compost are subject to self-degradation in the long-term thereby generating CH<sub>4</sub> rather 1229 than mitigating it <sup>[7]</sup>. Similarly, the physical processes such as capillary action may 1230 develop in some covers such as biocovers in the event of precipitation due to the stark 1231 differences in the hydraulic conductivities of biogenic layer and gas distribution layer 1232 causing occlusion of the pores and limiting gas transport. Hence, it necessitates further 1233 research in exploring alternative cover materials which have better long-term 1234 performance without significant economic ramifications as well as improve the cover 1235 1236 design to incorporate any potential water-logging due to infiltration. Slope stability: With the increasing scarcity of open lands, the landfills are designed with 1237 steeper slopes and greater heights to increase the waste containment capacity. This calls 1238 for developing cover systems with enhanced slope stability. It presents a bigger challenge 1239 for alternative cover systems whose core function is gas mitigation or infiltration barrier. 1240 Hence, it is a challenge to design alternative cover systems to provide slope stability in 1241 addition to gas mitigation and minimizing infiltration. 1242

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## 1245 6 Summary

Landfilling has transformed enormously over the years from mere dumping to modern engineered landfills. Landfill covers have evolved from basic soil covers protecting breeding of flies and birds to advanced engineered cover systems with impermeable GMs and gas collection

systems. The regulations regarding landfill cover management have become stricter over the 1249 decades to reduce leachate generation and prevent emissions of LFGs. The conventional landfill 1250 covers are mainly designed to prevent infiltration into the landfill to minimize the leachate 1251 generation. The newer regulations require landfills to manage LFG emissions by installing gas 1252 collection systems if the CH<sub>4</sub> emissions are high. The older landfills where providing gas 1253 1254 collection systems is not economically and practically feasible, managing gas migration becomes a major challenge. Similarly, conventional cover systems are cost intensive and are susceptible to 1255 desiccation cracking and erosion, leading to increased infiltration and gas migration. Hence, 1256 extensive research has been conducted in the past two decades exploring various alternative 1257 cover systems. 1258

The alternative cover systems are required to meet the regulatory requirements in terms of minimizing the infiltration. Various alternative cover systems such as ET covers, anisotropic covers, capillary barriers and engineered turf covers have been developed which utilizes the natural processes to control rainwater infiltration into the waste. These covers are not only economic but also add aesthetic value to the landfills. However, each of these cover systems suffer from some limitation regarding their applicability in dynamic climatic regions.

As MSW landfills emerged as a prime contributor of anthropogenic CH<sub>4</sub> missions globally, CH<sub>4</sub> oxidation potential of landfill cover soil was investigated, and several organic based alternative cover systems such as biocovers have been explored to enhance microbial CH<sub>4</sub> oxidation in landfill cover. Several organic materials have been tested as the biocover substrate such as compost, waste biocover, earthworm cast and sewage sludge. One of the challenges associated with using biobased cover systems is their long-term stability and hence there is a need to explore cover systems which are durable and sustainable at the same time. In this regard, biochar-based cover systems are gaining popularity as they are more recalcitrant under adverse
climatic conditions and enhances the physical properties of the cover systems favoring the
growth of CH<sub>4</sub> oxidizing microbial population.

Apart from CH<sub>4</sub>, other trace gas emissions such as H<sub>2</sub>S, methyl mercaptan, aldehydes, 1275 BTEX compounds, etc., from landfills pose serious concerns for health and environment. 1276 1277 Alternative cover systems have been explored to mitigate these non-methane emissions from landfills. However, there is a need to develop an integrated cover system which addresses all the 1278 fugitive LFG emissions together resulting in zero emissions at the landfills. The newly proposed 1279 biogeochemical cover system offers promise in mitigating three of the major LFG components 1280 (CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>S) by utilizing byproducts from the steel making process (steel slag) and waste 1281 biomass in the form of biochar. This could be an environment friendly and sustainable 1282 alternative to the fugitive LFG emissions however, the cover system is still in its infancy and 1283 needs to be validated with extensive field-testing programs to affirm the laboratory findings and 1284 conclusions. 1285

Altogether, this review outlines how MSW landfill cover systems have evolved over the past three decades and how these advancements in the cover systems are benefitting the environmental pollution prevention. Furthermore, this study also presents the basic mechanisms underlying the functioning of the various alternative cover systems for mitigating various components of LFG. In the end, the study casted light on the research challenges associated with the development and performance of the alternative cover systems and prospects for the researchers.

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# 13018Conflict of Interest Statement

1302 The authors have no conflict of interest.

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