CHAPTER 2

LITERATURE REVIEW

2.1 Waste

Coined as the by-product of human activity, waste is any material or product that has no value in the perception of the generator or the consumer (Agamuthu, 2001). In today's economy, waste plays a significant role with the ever increasing problem of sustainable waste management. Conversely, the amplified volume of solid waste in the past decades grounds from rapid population growth, urbanization, economic development, industrialisation and affluence (World Bank, 2012). Solid waste is commonly generated by human and animals which are normally solid and discarded. There are four main categories of solid waste; (1) MSW which includes household waste, commercial waste and institutional waste; (2) Hazardous waste which also includes clinical waste; (3) Agricultural waste and; (4) Industrial waste (Agamuthu, 2001). MSW is the largest contributor to the waste stream and the biggest problem faced by the growing urban cities.

2.2 Waste Generation

2.2.1 Global

The world population is expected to grow by one-third by 2030 in sync with the doubling of the world's economy. Higher income level and standard of living have paved the way to the rise in MSW generation in the recent decades. Statistics provided by the World Bank (2012), indicates that the current global MSW generation rate by its 3 billion residents is estimated at 1.3 billion tonnes per year and is expected to ascent to 2.2 billion tonnes with 4.6 billion residents by the year 2025. Countries from the Organisation for Economic Co-operation and Development (OECD, 2013) produced almost half of the world's waste, while Africa and South Asia regions produced the least waste. The table below shows the per capita generation of MSW in different countries and cities around the world (**Table 2.1**).

Table 2.1: Per capita generation of waste in different countries and cities

Country	**Waste generation (kg/percapita/day)**	**References**
United States of America (USA)	1.96	World Bank (2012)
Sweden	1.40	OECD (2008)
Germany	1.58	OECD (2008)
United Kingdom (UK)	1.54	OECD (2008)
Malaysia	1.30	Agamuthu & Tanaka (2014)
Hong Kong, China	1.33	Li *et al.* (2009)
Mexico	0.68	Couth & Trois (2010)
Chennai, India	0.60	Esakku *et al.* (2007)
Delhi, India	0.47	Esakku *et al.* (2007)
Sub-Saharan Africa	0.65	World Bank (2012)

From **Table 2.1** it is evident that MSW generation is a function of affluence with the per capita generation spanning a wide range, from 0.47 to 1.96 kg/capita/day following the economic status of the countries and cities.

2.2.2 Malaysia

In Malaysia, 65% of the total population currently lives in the cities and the urbanisation level has greatly influenced the MSW generation (Agamuthu *et al.*, 2009). In 1970's Malaysia recorded an urbanisation rate of 26.8% and the figure rose to 71.0% in 2010 (Compendium JPM, 2012). With rapid development of urban area, rural-urban migration, change in lifestyle and consumption pattern, the MSW generation has increased by more than 91% over the past decades (Agamuthu *et al*., 2009). The daily generation of waste escalated from 22,544 tonnes in 2003 to 33,000 in 2013 marking a 46% hike in just 10 years (Agamuthu & Tanaka, 2014; KPKT, 2013). **Table 2.2** shows the trend in waste generation in Malaysia by states from 2003 to 2012 (KPKT, 2013; Fauziah, 2009).

Table 2.2: Daily generation of MSW in Malaysia from 2003 to 2012 (KPKT, 2013)

Year / State	**2003**	**2006**	**2009**	**2012**
Johor	2199	2475	2549	3005
Kedah	1520	1711	1762	2078
Kelantan	1188	1337	1377	1623
Melaka	590	664	684	808
Negeri Sembilan	869	978	1007	1188
Pahang	1099	1236	1273	1502
Perak	1753	1973	2032	2397
Perlis	224	252	260	307
Pulau Pinang	1248	1405	1447	1707
Selangor	3245	3653	3763	4435
Terengganu	1013	1140	1174	1385
Wilayah Persekutuan • Kuala Lumpur • Putrajaya • Labuan	2965	3337	3437	3965
Sabah	2641	3002	3092	2137
Sarawak	1982	2210	2276	2028
Total	**22544**	**25372**	**26134**	**28565**

2.3 Waste composition

The MSW composition varies from one country to another and is the pre-determinant factor for sustainable waste disposal method. In Malaysia, organic waste dominates the waste stream by more than 40% and it mainly consists of kitchen waste and food waste (Agamuthu *et al*., 2009). Similar results were found from a study done in China by Yuan *et al.*, (2006), where the MSW was also dominated by waste with high organic and moisture content and makes up to approximately 60% of the total MSW. However, this is in contrast with many developed and industrialised countries who's waste consist more of recyclable and bio-degradable organic waste (Zhang *et al.*, 2010). For example in Western Europe, the waste composition features a high percentage of paper and plastic (Zhang *et al.,* 2010). From the World Bank report (2012), it is further deduced that the low income countries make up for 64% and 5% of organics and paper in their MSW whereas in the high income countries it is 28% and 31%, respectively. Higher income countries have applied many techniques to tailor suite its own unique waste management system. This is to manage various components of waste streams in a flexible manner. **Figure 2.1** shows the waste composition in countries with different income level with comparison to Malaysia. Malaysia, which is categorised as a middle income country, has lower amount of organic waste compared to the statistics of the middle income country. However, in Malaysia the plastic waste is still mushrooming due to lack of awareness and NIMBY attitude by many manufacturers and end users.

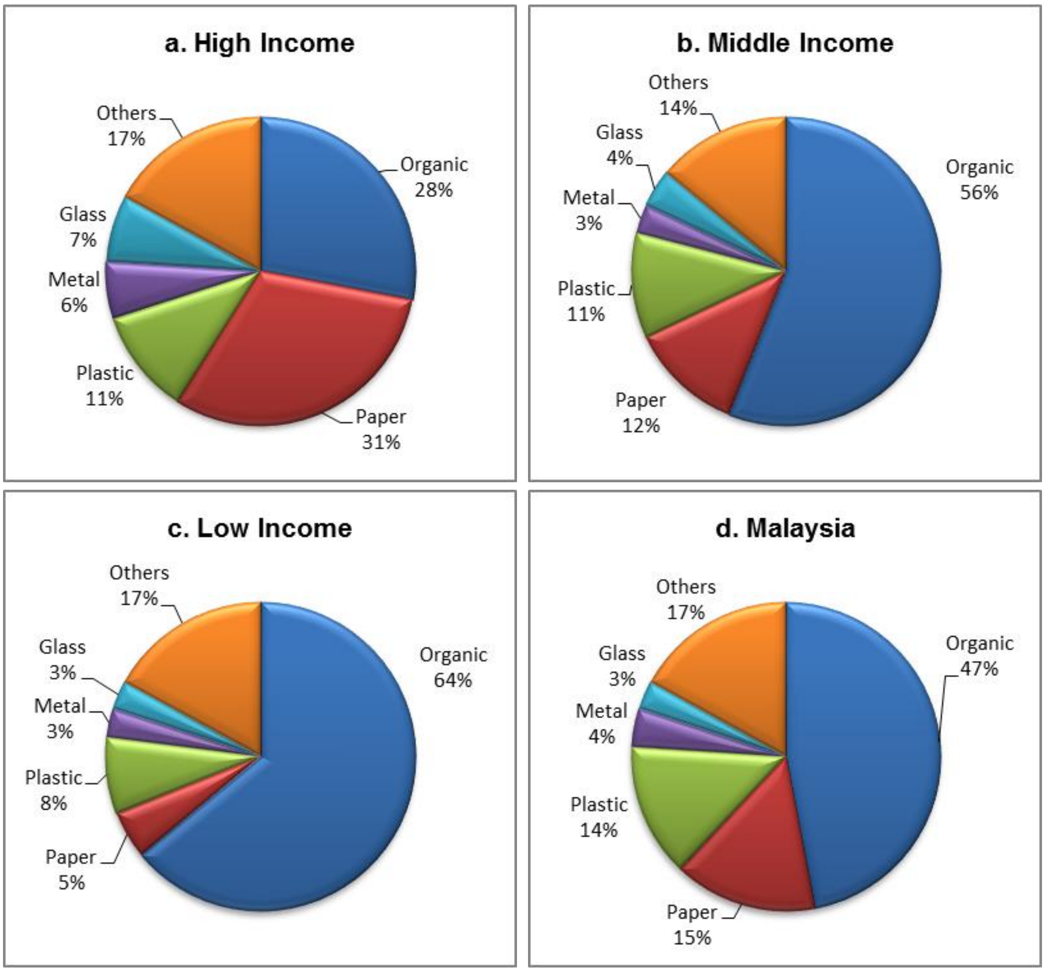

Figure 2.1: Waste composition by Income level (a, b, c) and in Malaysia (d) (*KPKT, 2013 and World Bank, 2012)*

2.4 Solid waste management

The need for sustainable waste disposal operation within a country has a ripple effect as it drives legislation, action by the authorities and new technical and technological innovations (Agamuthu *et al*., 2009). In the European Union (EU) countries, comprehensive policies and regulations regarding waste management were introduced as early as the 1990's. The Landfill Directive of 1999 by EU, limits the amount of waste going to the landfill and newly constructed landfill must be equipped with liners and gas capture systems (ISWA, 2009). However, in majority of developing countries the development of modern and sustainable system of waste management is frequently delayed due to the scarcity in policy implementation, political set-up, as well as, for some technical and economic reasons. A similar scenario was observed in Malaysia. After a 10 year debate on the solid waste bill, the Government of Malaysia approved The Solid Waste & Public Cleansing Management (SWPCM) Act in 2007. The Act transferred the responsibility of solid waste management from the state government and local authorities (LA's) to the Federal Government to implement an integrated solid waste management system (Agamuthu *et al.*, 2009). This Act is expected to bring new challenges and major changes in solid waste management in Malaysia.

2.5 Waste management practices

A wide range of waste management methods have been practiced to address the growing demand in waste disposal. Many factors like technology advancement, economic sustainability, policies, environmental and health benefits, waste capacity, waste composition and climate conditions are considered in choosing the best waste management practices. At the same time, the general perception that waste management still pose considerable environmental threats is inevitable although waste management practices have evolved significantly over the last decades (Polettini, 2012).

The waste management options practiced are often classified in a waste management hierarchy following an order of the least impactful to the environment to the most (Raina, 2010). The waste management hierarchy’s usage dated back to 1970’s during the Ontario’s Pollution Probe (World Bank, 2012) and started with the use of 3R’s (reduce, reuse, recycle), as well as, the edition of the latest R – recovery. In this hierarchy, waste prevention or reduction is given the highest priority while disposal methods such as landfilling and incineration, are the least preferred.

Waste prevention and reduction refers to reducing waste at the source which includes reusing products, donating items, buying in bulk, redesigning products and reducing packaging. The recycling and composting practice focuses on activities such as collecting, sorting and processing the recyclable products as well as composting of food waste and yard trimmings. In energy recovery the conversion of non-recyclable waste material into heat, electricity and fuel through many waste-to-energy (WTE) processes. Although landfilling and incineration are the most widely used disposal method globally, it is still the least preferred waste management practice due to its environmental impacts (USEPA, 2012). **Figure 2.2** shows the waste management hierarchy in an inverse triangle image. The larger the portion of the triangle, the higher the priority that is given to that waste management practice.

Figure 2.2: The waste management hierarchy (USEPA, 2012)

Figure 2.3 shows the common waste management practices in few countries around the world in year 2008 (OECD, 2008). In Germany, 47% of the MSW was recycled while another 32% was incinerated. Only 0.6% of the MSW was sent directly to the landfills. In countries like France, Italy, Japan, UK and the USA, recycling rates were recorded from 17 to 25% (OECD, 2008). Stringent legislation, regulations and policies such EU directives and Zero Organic Waste to Landfills Policy have paved the way to the more environmentally sound strategies for MSW disposal in most of the EU countries (ISWA, 2009). While in Japan, land scarcity has made them depend on incinerators to dispose their waste. However even with many environmentally sound waste management options, landfilling still remains as the main option for MSW disposal in many countries. In countries like the USA, UK, China, Italy and many other developing nations, landfills are the final waste disposal sites as it is the cheapest and easiest method to adhere (SivaShangari & Agamuthu, 2013; OECD, 2008).

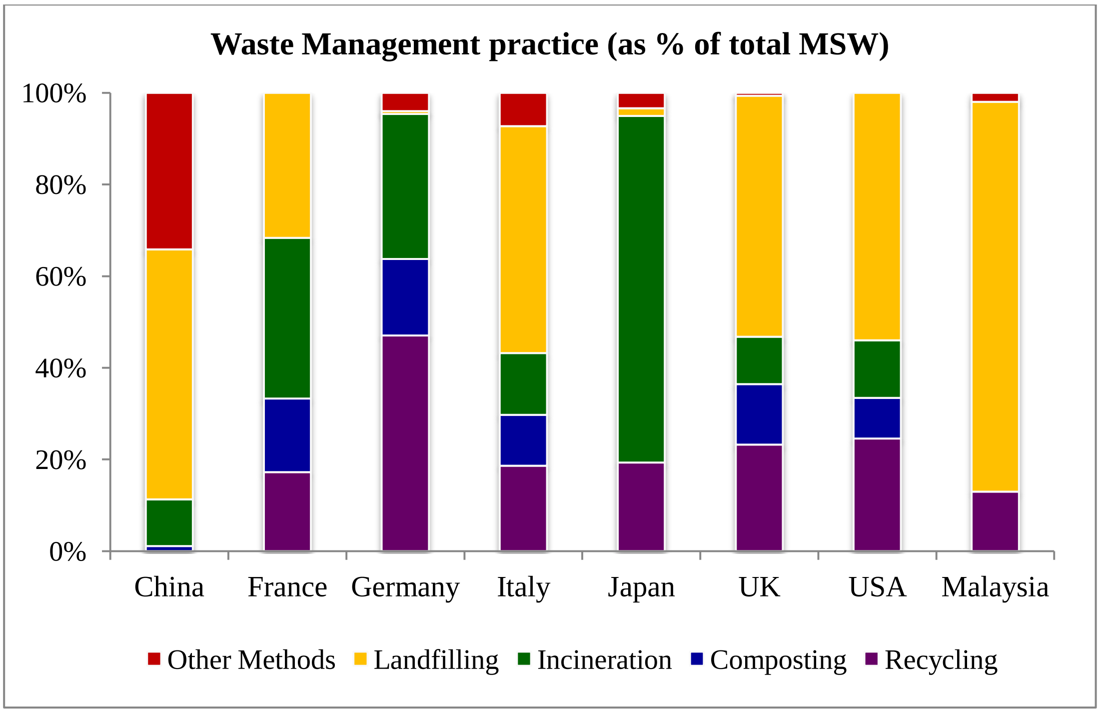

Figure 2.3: Waste management practices in various countries (SivaShangari & Agamuthu, 2013; OECD, 2008)

2.6 Climate change and waste

Climate change has become a hotly debated global issue. The increase in GHGs emission and global warming has been given the utmost importance to understand, mitigate and adapt to climatic changes occurring around the world. Also, the initiation of the International Treaty of Kyoto Protocol in 1997 and its enforcement in 2005, established a worldwide cooperation in GHGs reduction (UNFCCC, 2012). Following this was other important climate negotiations which outlined action plans like The Bali Road Map in 2007, Copenhagen Accord in 2009, Cancun Agreement in 2011, Durban Outcomes in 2012 and the latest Doha Climate Gateway in 2013 (UNFCCC, 2014). All these talks and action plans were targeted at reducing GHGs emissions and turning down the projected global temperature increase of 4˚C to only 2˚C climate goal (UNFCCC, 2012; ECOFYS, 2010). The industrialised countries or popularly known as the Annex 1 countries, as well as, the seven major emerging economies of non-Annex 1 countries (Mexico, Brazil, China, India, Indonesia, Korea and South Africa) have pledged to reduce their GHGs emissions between 17 to 40% as compared to the year 2005 or their business as usual (BAU) level (COP15, 2009; ECOFYS, 2010). Malaysia too, in the Global Climate Change Summit (COP15), had pledged to voluntarily reduce CO_2 intensity up to 40% by 2020 as compared to 2005 levels (COP15, 2009). This had subsequently paved the way to the formulation of the National Green Technology Policy in 2011 which focused on energy, buildings, waste and transportation (KeTTHA, 2013).

As for the GHG emissions from the waste sector, it only denotes to 3-5% of the total global GHG emissions (World Bank, 2012; Bogner *et al.*, 2007). Although the quantity seems trivial, there is now credible evidence that, taking into account the associated avoided emissions, the waste sector can move from being a minor source of emitter to becoming a major saver of emissions (UNEP, 2010). It is a known fact that every waste management practice generates GHG, both directly from emissions and indirectly through energy consumption. Nevertheless, the overall climate benefit or impact of the waste management practices will depend on net GHGs, accounting for both emissions and indirect, downstream GHG savings. **Table 2.3** depicts the results by Polettini (2012) on several waste management practices and its GHG emissions and savings.

Table 2.3: Waste management practices and GHG emissions and savings adapted from (Polettini, 2012, Gentil *et al.*, 2009)

WM Practices	**Indirect (upstream)**	**Direct**	**Indirect (downstream)**
Waste reduction, reuse, recycling	E: Fuel used in the facility E: Consumption of heat and electricity E: Materials used in facility construction E: Transportation of materials to the facility	E: GHGs from fuel combustion	S: Avoided emissions from substituted raw materials (-390 kg CO_2-eq tonne^{-1} ww observed in Germany) S: Avoided emissions from avoided use of harvested wood E: Transportation of recovered materials for reuse/recycling E: Transportation of residues for treatment/final disposal
Mechanical biological treatment	E: Fuel used in the facility E: Consumption of heat and electricity E: Materials used in facility construction E: Transportation of materials to the facility	E: GHGs from fuel combustion E: Constituents of biogas from waste degradation (aerobic/ anaerobic)	S: Avoided emissions from biogas use for energetic purposes (-103 kg CO_2-eq tonne^{-1} ww observed in Germany) S: Avoided emissions from materials recycling S: Avoided emissions from compost use as a substitute of soil growth media S: Carbon storage in compost E: Treatment of the off-gases and leachate E: Transportation of recovered materials for recycling E: Transportation of residues for treatment/disposal
Thermal treatment	E: Fuel used in the facility E: Consumption of heat and electricity E: Materials used in facility construction E: Transportation of materials to the facility	E: GHGs from thermal waste conversion	S: Heat/electricity from substituted fossil fuels (-563 kg CO_2-eq tonne^{-1} ww observed in Dermark) S: Avoided emissions from recovered metals from the ash S: Avoided emissions from use of ash as a substitute of natural aggregates E: Transportation of recovered materials for recycling E: Treatment of residual ashes E: Transportation of residues for treatment/final disposal S: Carbon sequestration by residual ashes
Traditional landfilling	E: Fuel & energy used at the landfill site E: Materials used in landfill construction E: Transportation of materials to the facility	E: Constituents of LFG from waste degradation (+454 and +346 kg CO_2-eq tonne^{-1} ww in Greece and Poland) E: GHGs from fuel combustion E: Emissions from leachate treatment	S: Avoided emissions from LFG combustion S: Carbon storage within the landfill body S: Advanced CH_4 oxidation in active covers

*Footnote : **E** – Emission ; **S** – Savings, (+) = Emission and (-) = savings, ww – wet waste*

Currently, the OECD countries generate the highest levels of methane. However, the developing nations are anticipated to increase significantly as better waste management practices lead to more anaerobic, methane producing conditions in landfills (UNEP, 2010). Landfilling especially in developing countries like Asia, South America and Africa are reported to contribute about 37 million tonnes of CO_2-eq or 40% of the annual CH_4 emission (Bogner *et. al.*, 2007). Global CH_4 budget from year 2000 to 2009 for agriculture and waste was between 187 to 224 $Tg(CH_4)\ y^{-1}$. These anthropogenic emissions are from paddy agriculture, ruminant livestock, landfills, man-made lakes and wetlands, and waste treatment (IPCC, 2013).

2.7 Landfilling as a MSW disposal option

Landfilling is the most widely used and economically viable solution for final disposal of MSW in many countries around the world. High cost of treatment and comparative disposal alternatives are a major reason for the reliance on MSW landfilling particularly in developing countries (Laner *et al.*, 2012; Brunner & Fellner, 2007). Similarly in Malaysia, 85% of the daily collected MSW is landfilled while only 15% are recycled (Agamuthu *et al.*, 2009). Based on a report by the Malaysian Ministry of Urban Wellbeing, Housing and Local Government (KPKT, 2012), there are a total of 296 landfills in Malaysia in 2012 with 56% of landfill in Malaysia are currently in operation (**Table 2.4**). Out of this total, only 8 are sanitary landfills equipped with adequate high density polyethylene (HDPE) liner, gas capture wells and leachate treatment ponds. The rest of it are common landfills or mere dumpsites with could pose great danger to the adjacent residents or water supply.

Table 2.4: Total landfills in Malaysia according to the states (KPKT, 2012).

State	Operational landfills	Non-operational landfills	Total
Johor	14	23	37
Kedah	8	7	15
Kelantan	13	6	19
Melaka	2	5	7
Negeri Sembilan	7	11	18
Pahang	16	16	32
Perak	17	12	29
Perlis	1	1	2
Pulau Pinang	2	1	3
Sabah	19	2	21
Sarawak	49	14	63
Selangor	8	14	22
Terengganu	8	12	20
WP Kuala Lumpur	0	7	7
WP Labuan	1	0	1
Total	**165**	**131**	**296**

2.7.1 Non-sanitary landfills and dumpsites

Non-sanitary landfills or also known as tip, dumpsites and dumping ground are in abundance around the world. These landfills are often used as burial areas for waste and is closed with some soil when the height of waste reaches a desirable maximum at the site (Themelis & Ulloa, 2007). These types of landfills are neither engineered with any liner system, leachate treatment ponds nor gas collection facility and pollute the local environment through bad odour and ground and surface water contamination. In a study by Manfredi *et al.*, (2009), open dumpsites in European countries produce 1.00 tCO_2-eq of LFG for per tonne of wet waste.

2.7.2 Sanitary landfill

Sanitary landfills are modern landfills which are well-engineered, designed, operated, monitored and located away from residential areas. These landfills are constructed in accordance with the government regulations to ensure the safety of people and the environment. Among the common regulations administered for a sanitary landfill are location restriction, artificial liner requirements, leachate collection and removal systems, gas collection and control system, groundwater monitoring system, as well as, closure and post closure requirement for the landfill (USEPA, 2012). Barton *et al*., (2008), concluded that sanitary landfills in developing countries could produce up to 1.48 tCO_2–eq of LFG for per tonne of wet waste. **Figure 2.4** shows a sanitary landfill.

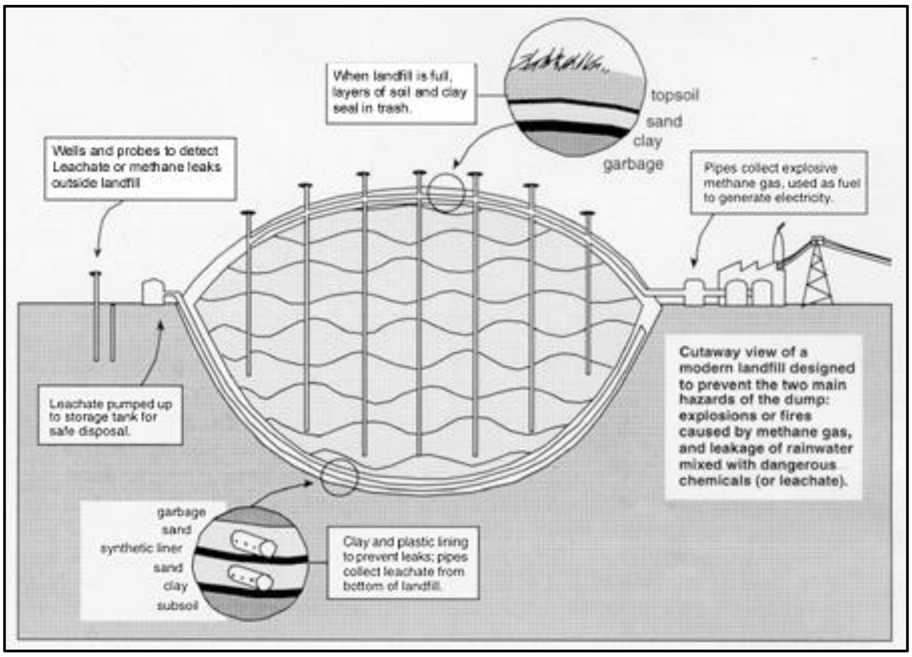

Figure 2.4: A sanitary landfill (USEPA, 2012)

2.8 Landfill Output

The organic compounds in the MSW are converted to gaseous or liquid states by undergoing various chemical and biological transformation (Wang *et al.*, 2012). The decomposition of MSW with the presence of rain water produces landfill gas (LFG), leachate and other inert solid waste as the primary output. Organics in the solid waste decomposes to produce LFG which contains CH_4, CO_2 and other trace amounts of toxic substances (Park & Shin, 2001). Conversely, landfills are also known as long-term sink for carbon sources (Bogner *et al.*, 2007). Previous studies by Bogner (1992) and Barlaz (1998) estimates that a minimum of 50% of the organics landfilled is not converted to LFG but still remains in the landfill. This condition is due to the presence of inert organics, slowly degrading cellulose and hemicellulose and biologically recalcitrant (lignin) constituents within the MSW landfilled (Polettini, 2012; Bogner *et al.*, 2007).

2.9 Landfill gas

2.9.1 Landfill gas generation

The MSW buried in landfills are subjected to physical, chemical and biological processes in which the biodegradable fractions decompose in an anaerobic condition through a complex series of microbial and abiotic reactions (Wangyao *et al.*, 2010). Accordingly, the biodegradation of organic waste takes place in five stages and leads to the production of landfill gas (LFG). The 5 stages are (1) aerobic (hydrolysis and aerobic degradation converting ready degradable carbohydrates to simple sugars, carbon dioxide (CO_2) and water (H_2O)); (2) acidogenic (hydrolysis and fermentation of simple sugars to solute volatile acids); (3) acetogenic (conversion of soluble acids to acetic acids, hydrogen (H_2) and CO_2); (4) methanogenic (methane (CH_4) generation bacteria metabolizes acetate and forms CH_4 and CO_2 – LFG); (5) aerobic (oxidation of CH_4 with the re-establishment of aerobic conditions)(Couth *et al.*, 2011; Environment Agency, 2004). The stages above can be simplified in a formula to represent the maximum amount of LFG generation as below (Nickolas & Priscilla, 2007):

$$C_6H_{10}O_4 + 1.5H_2O = 3.25CH_4 + 2.75CO_2$$

Figure 2.5 shows the flow chart of the process.

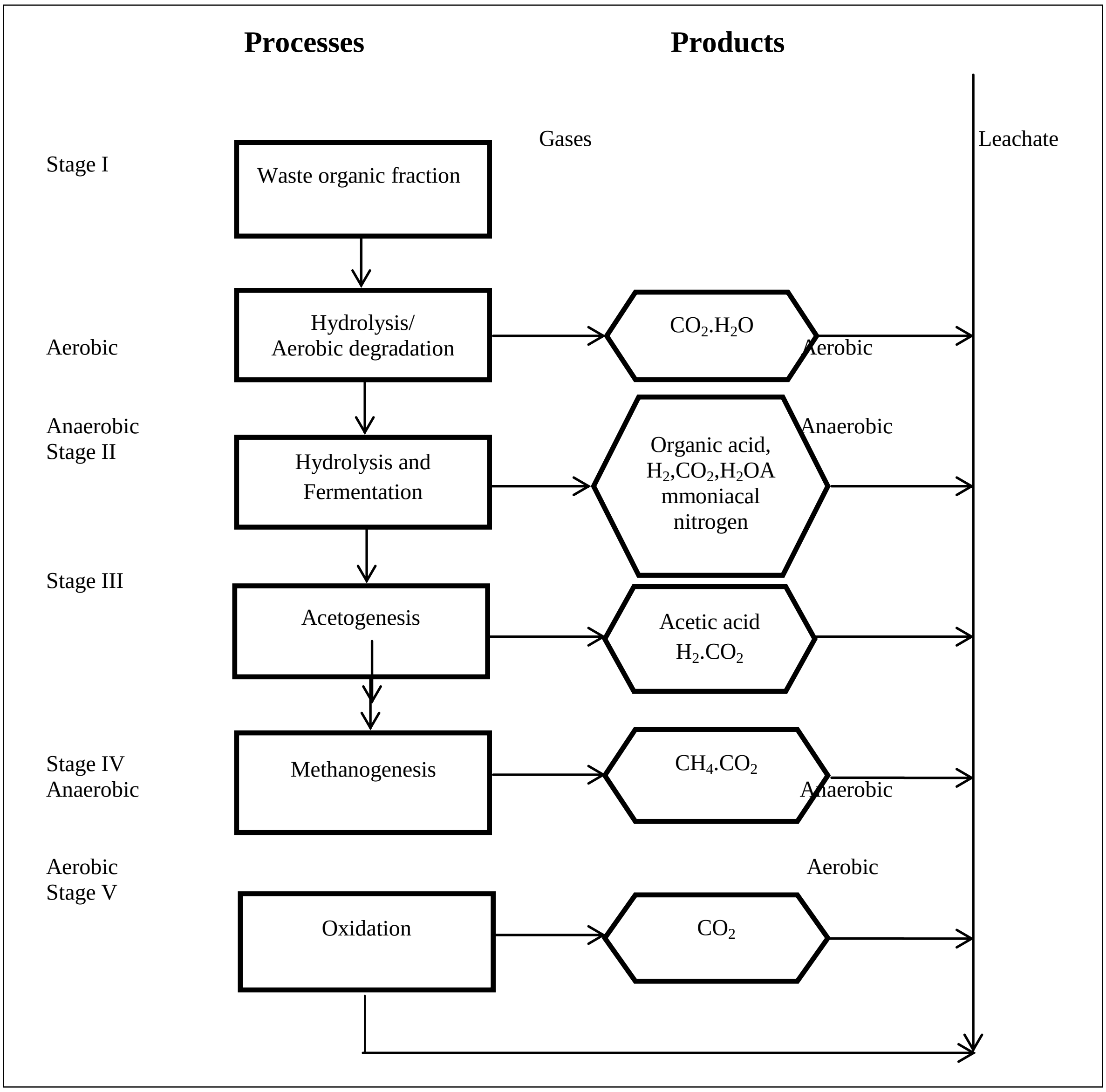

Figure 2.5: The major stages of waste degradation in landfills (Williams, 2005)

2.9.2 LFG Composition

Abichou *et al.,*(2004) and Humer & Lechner (1999) denoted that the biological decomposition of one tonne of MSW will produce approximately 160 m^3 to 250m^3 of LFG. LFG is composed by a mixture of gases compound as shown in **Table 2.5**. In general more than 50% of the LFG is CH_4, followed by CO_2 between 40 to 60% and other trace amounts of various chemical compounds (Hegde *et al.*, 2003). Although CO_2 is produced in landfill, it is said to have neutral greenhouse effect due to its biogenic origin. Its generation in a landfill is also negligible compared to other CO_2 producing sectors such as industry, energy and transportation (Polettini, 2012).

Table 2.5: The composition and characteristics of LFG (ATSDR, 2001; Tchobanoglous *et al.*, 1993)

Component	**Volume (%)**	**Characteristics**
CH_4	45 - 60	Is colourless, odourless and naturally produced gas.
CO_2	40 - 60	Is colourless, odourless and slightly acidic.
Nitrogen (N_2)	2 - 5	Is colourless, odourless and tasteless. The major contributor to atmospheric gas (79%).
Oxygen(O_2)	0.1 - 1	Is colourless, odourless and tasteless. The second major contributor to atmospheric gas (21%).
Ammonia (NH_3)	0.1 - 1	Is a colourless gas with a pungent odour.
Non-methane organic compounds (NMOCs)	0.01 – 0.6	Is an organic compound that forms naturally or through synthetic chemical processes.
Sulphides	0 - 1	Is a naturally occurring gas with a smell like rotten-egg.
Hydrogen (H_2)	0 – 0.2	Is an odourless and colourless gas.
Carbon Monoxide	0 – 0.2	Is an odourless and colourless gas.

Inversely, CH_4 is a more significant GHG in the landfill. Given a time horizon of 100 years, CH_4 has a global warming potential of 25 which means it is capable of absorbing and trapping infrared radiation 25 times more as compared to CO_2 and has a longer residence time in the atmosphere (Pachauri & Reisinger, 2007). Globally CH_4 from landfills represent 12% of the total anthropogenic CH_4 emission (World Bank, 2012) and the estimated release is between 35 and 69 Tg (CH_4) y^{-1} (Bogner *et al.*, 2007). In a separate report by IPCC (2013), landfills are responsible for almost half of the CH_4 emissions attributed by the MSW in 2010. In Malaysia, where almost 85% of the daily generated MSW is landfilled, 47% of national CH_4 average is from landfills (Agamuthu & Fauziah, 2012; NC2-Malaysia, 2010).

2.9.3 Factors Influencing gas generation

The rate and volume of the LFG generated in landfills are influenced by several factors. According to Agamuthu (2001), the rates of LFG production depend on size and composition of solid waste, age of solid waste and landfill, moisture content of waste, temperature conditions in landfill, quantity and quality of nutrients, organic content of refuse, pH and alkalinity of liquids in the landfill and presence of toxic and hazardous materials in landfills. Some of the factors are described further below:

a) **Waste composition**: The bacteria within the landfills feed on the organic matter while decomposing. The optimum ratio of organic matter (expressed as chemical oxygen demand (COD), nitrogen (N) and phosphorous (P) is 100:0.44:0.08 (Christensen *et al.*, 1996). In addition, nutrients and metal supplements have a positive effect on the LFG production rate (Isci & Demier, 2007). Gas generation is also increased by the presence of putrescible materials (Christensen *et al.*, 1996).

b) **Age of refuse**: waste buried in the landfill within the recent years (less than 10 years). Peak gas production occurs between 5 to 7 years after the waste is buried. However, small quantities of LFG may continuously be emitted from a closed landfill for 50 or more years (ATSDR, 2001).

c) **Presence of oxygen**: O_2 is considered as inhibiting for CH_4 formation. The absence of O_2 is important for the anaerobic bacteria to grow and convert solid carbon to CH_4 and CO_2 (Christensen *et al.*, 1996).

d) **Moisture content**: Rees (1980) had summarized that the LFG production rate increases between 25% to 60% water content. The increase of water content limits O_2 transport and facilitates CH_4 production. However, saturated moisture content hinders CH_4 production (Christensen *et al.*, 1996; Rees, 1980). While a moisture content of 40% or higher based on wet waste, promotes maximum gas production (ATSDR, 2001) .

e) **Temperature**: Like many other microbial processes, CH_4 formation is highly affected by temperature. The warmer the temperature, the higher the bacterial activity which in turn increases the rate of LFG production. At elevated temperatures, the CH_4 production is more rigorous and produces more heat, as such is self-enhancing. During the decomposition process, the bacterial releases heat which stabilizes the temperature of between 25˚C to 45˚C (ATSDR, 2001).

f) **pH**: The methanogenic bacteria which produces CH_4 as a metabolic by-product, operates efficiently only within a narrow pH range of 6 to 8 (Abushamamala *et al.*, 2009). The methanogenic ecosystem in the landfill is rather delicate, and a balanced relation between bacterial groups is crucial for good CH_4 production (Christensen *et al.*, 1996).

2.10 Impact of landfill gas emission

LFG contains H_2 and CO_2 in the initial phase of LFG production from the waste degradation process but at the final stage it is mainly a mixture of CH_4 and CO_2 (Williams, 2005). All these gases have generated several impacts to the environment as well as human. The impacts are felt at different scales of radius depending on its severity as shown in **Figure 2.6**.

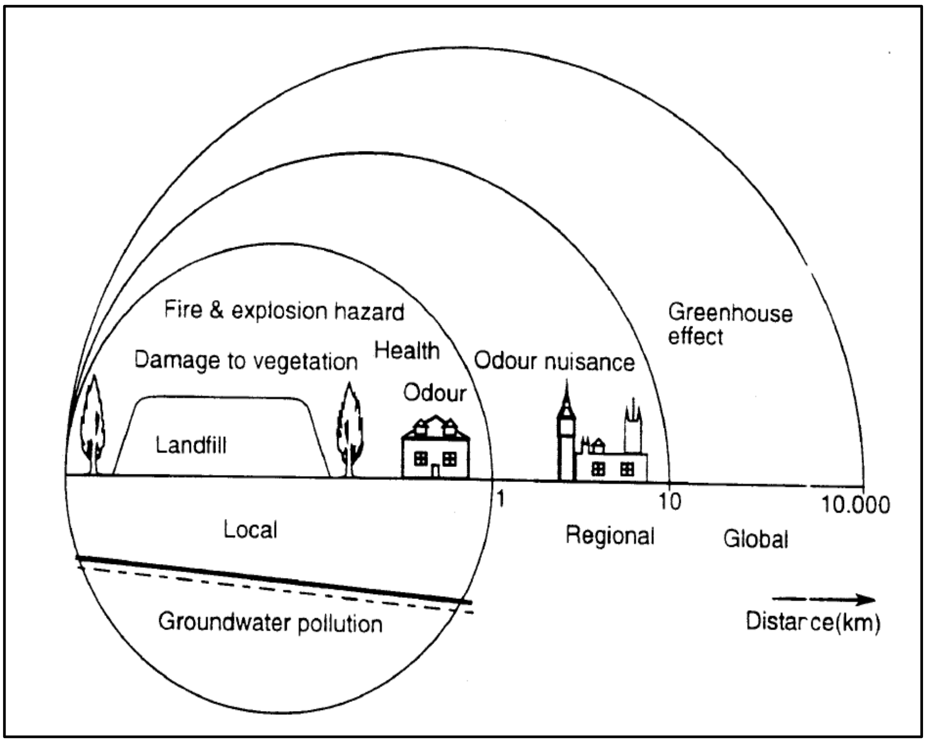

Figure 2.6: Different scales of landfill effects (Scheutz *et al.*, 2009; Luning & Tent, 1993)

2.10.1 Greenhouse effect and global warming

At the end of the 20th century and during the industrial era, the concentration of the global GHG(s) such as CO_2, CH_4 and N_2O showed major hikes. The CH_4 concentration generally showed an increasing trend since pre-industrial years (**Figure 2.7a**). But a more comprehensive derivation, magnified a decreasing growth rate of CH_4 from early 1980 to 1998, a stable growth rate period between 1999 and 2006 and an increasing growth rate thereafter from 2007 to 2011 **(Figure 2.7b)** (IPCC, 2013; Dlugokenly *et al.*, 2009; Rigby *et al.*, 2008). The current contribution of CH_4 to climate change forcing is 18% of the total radiative forcing by all long-lived GHG

(Foster *et al.*, 2007). Radiative forcing denotes to a measure of energy balance of the Earth-atmosphere system when factors that affect the climate are altered naturally or by human.

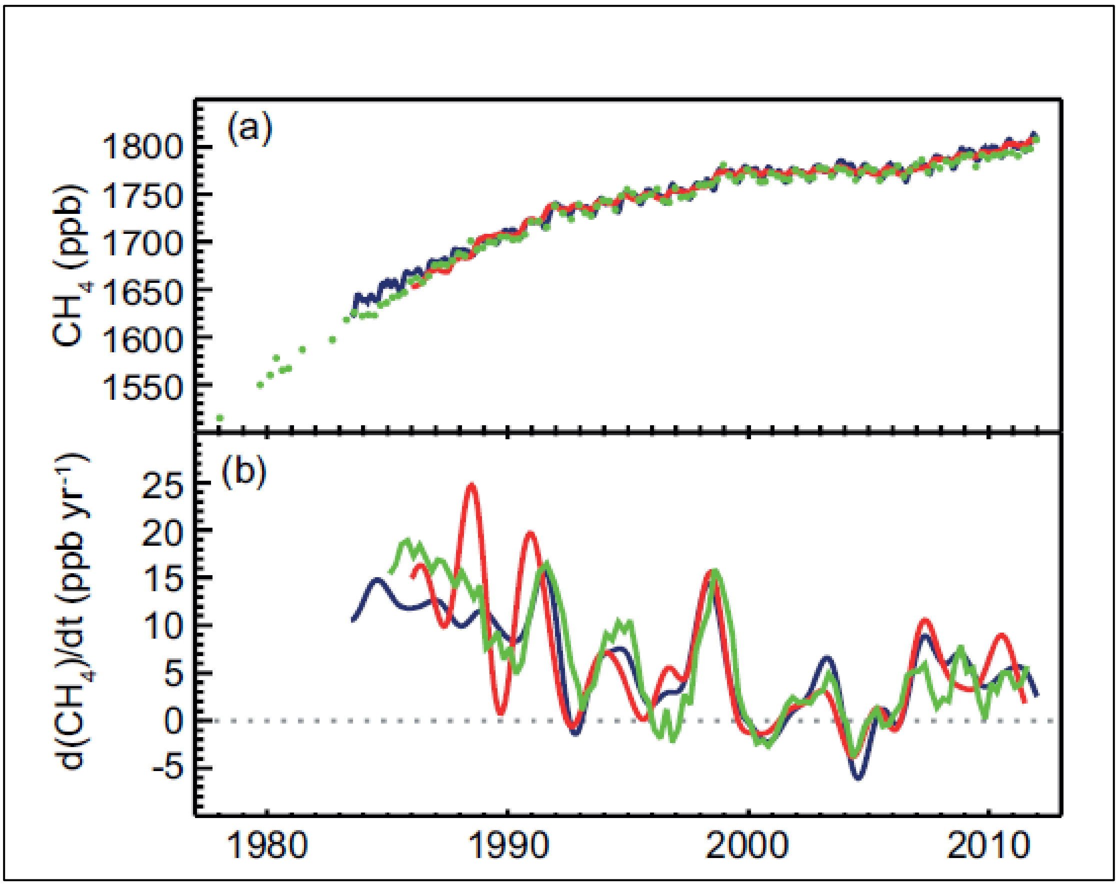

Figure 2.7: Global average of CH_4 from 1978 to 2011 and **Figure 2.8:** Instantaneous growth rate of globally averaged atmospheric CH_4 for the same period, line colours represents: **green**-quarterly average, **red**-monthly average, **blue**-quasi weekly average (IPCC, 2013; Dlugokenly *et al.*, 2009; Rigby *et al.*, 2008)

In a report by World Bank (2012), the GHG emissions from MSW have emerged as a major concern as post-consumer waste is estimated to generate almost 5% that is about 1460 metric ton CO_2-eq of the total global GHG emissions. Whereas, the global CH_4 emissions from landfills are estimated to be between 500 to 800 metric ton CO_2-eq per year (USEPA, 2006; Monni *et al*, 2006; Bogner & Mattews, 2003).

2.10.2 Odour and weather impact

The odour from landfills caused by secondary emissions is a growing concern to the area surrounding the landfill. Since both CH_4 and CO_2 are odourless, the stench smells from the landfills often originate from other principal components of LFG such as H_2S and organic sulphur compounds (mercaptanes) (Sitiaishah, 2011). Similarly in a study by Bertacchi *et al.* (1997), the main cause of odour in an Italian landfill was identified as dimethyl sulphide and carbon sulphite. In a case study done at Bisasar Road Landfill in Durban, South Africa, the meteorological parameter such as temperature, relative humidity and rainfall were evaluated against the odour complaint by the surrounding residents. The number of complains had increased during October to January (summer months) when the weather condition was hot, humid and wet (Stretch *et. al.*, 2001). These results are shown in **Figure 2.9**.

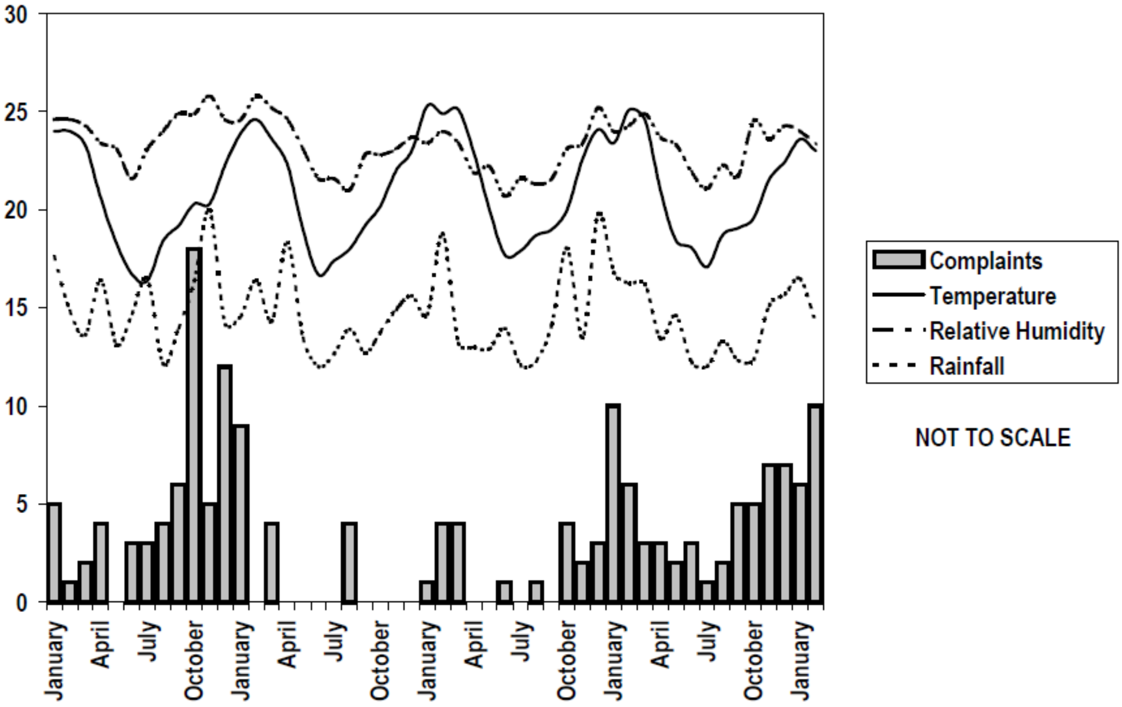

Figure 2.9: Temporal distribution of odour complaints and selected weather conditions from January 1997 to February 2001 at Bisasar Road Landfill. Weather data is not to scale (Stretch *et al.*, 2001)

2.10.3 Groundwater pollution

Landfill gas (LFG) migrating to the unsaturated zone surrounding the landfill is exposed to infiltrating water. The infiltrating of water is caused by rainfall, water present in the waste itself and water from biodegradation of waste. CO_2 and CH_4 are water-soluble (the solubility of CO_2 is 2320 mg l^{-1} and only 30 mg l^{-1} for CH_4 at 10˚C). Besides LFG, leachate produced by the landfill also contains large amount of contaminants such as heavy metals which are likely to cause groundwater pollution. In a study by Bundela *et al.*, (2012) in Jabalpur, India, the physico-chemical analysis of groundwater adjacent to the landfill found that the total dissolved solids (TDS) varied between 546 mg/L to 907 mg/L much higher than the permissible level of India. Therefore an engineered landfill with sound leachate removal and treatment pond will be the best option to cater this sort of problem.

2.11 Landfill CH_4 mass balance, controlling processes and factors

The landfill CH_4 mass balance and quantity of CH_4 emitted from landfills are affected by many different processes. It is important that the mass balance must be well understood before a landfill is designed and properly engineered. The processes of CH_4 mass balance are summarized in **Figure 2.10** which shows the migration of CH_4 through lateral direction, recovery and emission.

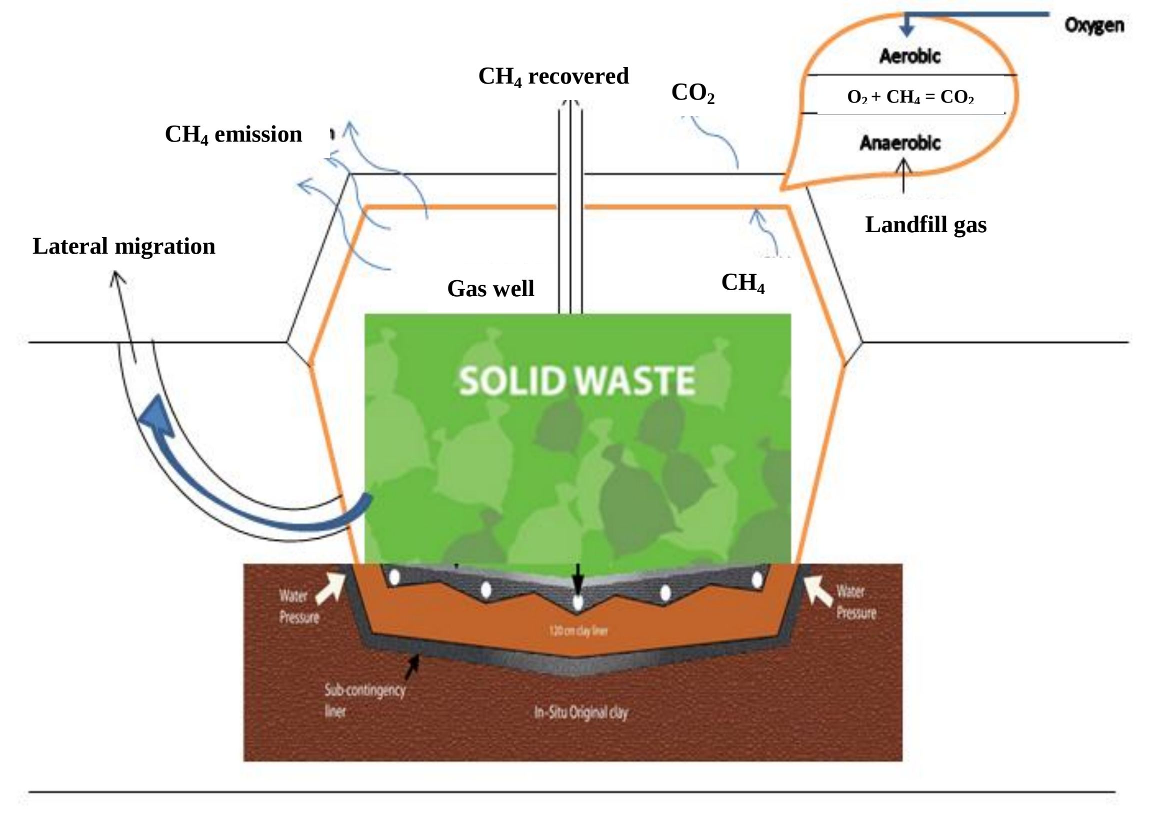

Figure 2.10: Landfill CH_4 balance

In an earlier study done by Bogner & Spokas (1993) CH_4 mass balance in the landfills was expressed by the following equation:

$$CH_4\ generated = \sum (CH_4\ recovery + CH_4\ emitted + CH_4\ lateral\ migration + CH_4\ oxidized) + \Delta\, CH_4\ storage \qquad \text{(all units = mass t}^{-1}\text{)}$$

The CH_4 mass balance associates CH_4 generation to CH_4 transport and microbial CH_4 oxidation terms. Lateral migration is usually qualitatively monitored at most landfill sites to some extent using gas probes placed around the landfill perimeter. The factors affecting the transport processes of LFG leading to gas emission or migration in a landfill are summarized in **Table 2.6**. Both ambient and internal factors greatly influence the LFG transfer processes. Therefore it is well documented that landfill CH_4 emission and migration is controlled by a combination of factors and processes.

Table 2.6: Factors affecting the LFG transport in a landfill (Scheutz *et al*., 2009)

Factors		
Meteorological conditions	**Soil/Color conditions**	**Waste & landfill conditions**
• Barometric pressure • Precipitation • Temperature • Wind	• Cracks • Permeability • Diffusivity • Porosity • Moisture content • Organic content • CH_4 oxidation capacity	• Gas production rate • Internal barriers • Gas vents • Lateral Migration Area

2.12 Quantification and estimation of LFG

LFG generation rates are currently estimated using mathematical models. The input parameters for these models are amount of waste landfilled for the calculated years and in most models also a specific composition of the waste. Most models calculates CH_4 generation potentials (L_o), degradable organic carbon (DOC), reaction constant (k) and subsequently the CH_4 generation released throughout the years. However, some models consider the degradation of waste over the years or decade while some do not. Among the models widely used are the IPCC model, the TNO-model, the

GasSim model, the LandGEM model, the Alvalzorg-model and the French E-PRTR model (Oonk, 2010).

2.12.1 The IPCC Waste Model

The LFG generation can be modelled empirically using zero-order, first-order or second-order generation model (Beh, 2013). However, studies have proved that higher order model have near accuracies to the measured data (Amini *et al.*, 2012; Oonk, 2010). Generally the first-order models are preferred as the second order models are more complicated and are not justified by increase in accuracy (Oonk *et al.*, 1994). The IPCC Model or better known as the IPCC Waste Model is based on a FOD method. In this model, it is assumed that the degradable organic component in the waste decays slowly for a few years or even decades. During this process, the LFG containing mainly CH_4 and CO_2 is formed. The rate of CH_4 production depends solely on the amount of carbon remaining in the waste provided the conditions within a landfill remains constant (IPCC, 2006). This model has a three tier hierarchy to calculate estimates of CH_4 emissions from solid waste disposal sites (Jensen & Pipatti, 2006). The higher tier with good quality data estimates more accurate results. The tiers are explained further in **Table 2.7**. The tier three calculations are used if country's specific data are available. The main uncertainties within the model arise due to the parameter used in this model rather than the method used.

Table 2.7 The 3 tiers of the IPCC Waste Model

Tier	Description
1	Estimation is based the IPCC FOD method using mainly default data and default parameter
2	Uses IPCC FOD method and some default parameter and historical waste data.
3	Methods are based on good quality country specific data and either uses FOD methods with i. Nationally developed key-parameter ii. Empirical country-specific parameter with key parameter like half-life and either methane generation potential (Lo) or DOC content in waste and the putrescible fraction of DOC (known as DOC_f)

This freeware model can be downloaded from the IPCC website and the main input parameters are amount of waste, and waste composition. This model is used to calculate the specific amount of CH_4 from the landfill and accommodates four different climate regions; wet boreal or temperate; dry boreal or temperate; wet tropical; and dry tropical. The climate region chosen determines the key parameters used within the model such as half-life, decay rate and degradable organic carbon. The equation used to calculate CH_4 emissions in this model is as follow:

$$CH_4\ emitted = \left[\sum CH_4\ generated - CH_4 recovered\right] \times (1 - CH_4\ oxidized)$$

Where the final amount of CH_4 emitted depends on the amount of CH_4 recovered and oxidized by the cover layer (IPCC, 2006). A number of studies have specifically focused on the use of this model which have been carried out in many countries to obtain a regional and national level CH_4 emission estimation (Penteado *et al.,* 2012; Abushammala *et al.*, 2012; Couth *et al.*, 2011; Wangyao *et al.*, 2010; Machado *et al.*, 2009; Weitz *et al.*, 2008, Jha *et al.*, 2008).

2.12.2 The TNO-Model

The Netherlands Organisation of Applied Scientific Research model or in short The TNO – model, describes the LFG generation as a function of amount of waste deposited from different organics (household waste, industrial waste, etc). The degraded organic carbon from waste is used to calculate the LFG produced. This single phase model was used by the Dutch to calculate and report the national averages of CH_4 emission. In studies done by Oonk *et al.* (1994) and Scharff & Jacobs (2006), CH_4 and CO_2 measurements from landfills were used to validate the TNO-model. Finally to calculate the CH_4 production, the LFG generated using the TNO-model is multiplied with CH_4 concentration of 50% and volumetric mass of 714g CH_4 m^{-3} (Scharff & Jacobs, 2006). The formula for the TNO-model is as follow (Thompson *et al.*, 2009):

$$Q = (DOC_f) \times (1.87) \times (M) \times (DOC) \times (k) \times (e^{(-kt)})$$

Where
Q is CH_4 production [kt/y]
DOC_f is the fraction of assimilated DOC
M is waste generation [Mt/y]
DOC is the degradable organic carbon [kg/tonne]
k is the decay rate [y^{-1}]
t is te time of waste disposal [y]
1.87 is the model multiplication factor

2.12.3 The GasSim model

The GasSim model was developed for the Environment Agency of England and Wales as a risk assessment tool to be used to evaluate the impact of LFG (Oonk, 2010). This model quantifies LFG related problems within a landfill, ranging from the CH_4 emissions, the effects of utilisation of LFG on local air quality, and LFG migration via subsoil to adjacent buildings (Oonk, 2010). The GasSim model does not provide the complete set of equations. Calculation modules within the program are protected from sharing which makes it difficult to be used without purchasing the software. At present, the LFG generated is based on the UK Waste Statistics. The principal input for this model is the amount of waste in megagrams (Mg), with the specific breakdown of the waste into different categories and carbon content (Scharff & Jacobs, 2006). The basic equation for this model is :

$$CH_4\, emission = CH_4\, production - CH_4\, recovery - CH_4\, oxidation$$

2.12.4 The LandGEM

The Landfill Gas Emission model or LandGEM was developed and widely used by the United States Environment Protection Agency (USEPA) to calculate the LFG emission from solid waste based on the USEPA standards. The model uses the mass of waste deposited and CH_4 generation capacity to determine the mass of CH_4 that is being generated. Mathematically LandGEM can be described using the following formula:

$$Q_{CH_4} = \sum_{i=1}^{n} kL_0M_i(e^{-kt})$$

Where

Q_{CH4} is CH_4 emission rate [$m^3CH_4y^{-1}$]

k is CH4 generation constant [y^{-1}]

L_0 is CH_4 generation potential [m^3CH_4.Mg waste^{-1}]

M_i is mass of waste in ith section [Mg]

T_i is age of the ith increment or section [y^{-1}]

Similar to the IPCC model, the LandGEM is based on the FOD method and can be used with the site specific data or default parameter in the absence of site-specific data (USEPA, 2004). This model too has been widely used by many researchers to quantify the amount of CH_4 generated in landfills (Sormunen *et al.*, 2013; Amini *et al.*, 2012; Bella *et al.,* 2011; Kamalan *et al.,*2011; Tolaymat *et al.*, 2010; Cruz *et al.*, 2010; Thompson *et al.*, 2009; Scharff & Jacobs, 2006).

2.12.5 The Afvalzorg – model

This model was developed by NV Afvalzorg in the Netherlands and is based on combination of literature and own experiences with landfill gas generation (Oonk, 2010). In this multi-phase model, eight waste categories and three fractions were distinguished (Scharff & Jacobs, 2006). For each fraction LFG production is calculated separately. The following equation describes the Afvalzorg model (Scharff & Jacobs, 2006):

$$\alpha_t = \varsigma \sum_{i=1}^{3} cAC_{0,i}k_{1,i}e^{-k_{1,i}t}$$

Where

α_t is LFG production at given time [m^3LFG y^{-1}]

ς dissimilation factor

i is waste fraction with degradation rate $k_{1,i}$ [$kg_i \cdot kg^{-1}$ waste]

c is conversion factor

A is amount of waste in place [Mg]

C_0 amount of organic matter in waste [kg per Mg of waste]

$k_{1,i}$ is degradation rate constant of fraction *i* [y^{-1}]

t is time elapsed since depositing [y]

2.12.6 The French E-PRTR or EPER model

The European Pollution Release and Transfer Register (E-PRTR) is the new Europe-wide register that provides easy access to environmental data from industrial facilities in the EU and few other places. This register replaces the previous European Pollutant Emission Register (EPER). Both the EPER and E-PRTR Protocols were used to determine the pollutant release in the air. This model gives two approaches to estimate CH_4 emissions from landfills; (i) by using the CH_4 emission estimates from LFG recovery in the landfill or (ii) by using a multiphase model (Oonk, 2010). The formula for this model is as follow (Kamalan *et. al.*, 2011; Oonk, 2010; Scharff & Jacobs, 2006):

$$FE_{CH4} = \sum_{x} FE_0 \times \left(\sum_{1,2,3} A_i \times p_i \times k_i \times e^{-k_i t} \right)$$

Where

FE_{CH4} is annual CH_4 production [m^3 CH_4 . y^{-1}]

FE_0 is CH_4 generation potential [m^3 CH_4 . Mg waste^{-1}]

p_i is waste fraction with degradation rate k_i [kg_i . kg^{-1}]

k_i is degradation rate of fraction i [y^{-1}]

t is age of waste [y]

A_i is normalization factor

2.12.7 Comparison of models for CH_4 generation

All the models above are compared using the CH_4 generation potential as shown in **Table 2.8**.

Table 2.8 Comparisons of models for CH_4 generation potential, L_0 (kg CH_4 per tonne waste) and half-lives for biodegradation for household waste (HHW) or MSW (Oonk, 2010)

Model	L_0 (kg/tonne)	Half-life (year)	remark
IPCC-model	63	12-23 (slow) 7 (moderate) 4 (fast degradable)	MSW Europe
TNO-model	60	7	Dutch HHW
GasSim	51	15 (slow) 9 (moderate) 6 (fast degradable)	UK HHW
LandGEM	122 72	14 (conventional) 35 (arid)	MSW USA
Afvalzorg	39-45	23 (slow) 7 (moderate) 3 (fast degradable)	Dutch HHW
E-PRTR (French)	55	5-10	France HHW

2.13 Validation of Model Performance

Models used to estimate the amount of LFG especially CH_4 emission always needs to be verified and validated for its persistency and accuracy. The comparison between observations and predicted CH_4 emission concentration was performed by applying statistical measures of error. The measures used reflected different aspect of spatial distribution of mean concentration and the characteristics of the concentration field (Cervone & Franzese, 2010). The error function analyses (EFA) that are commonly used are the fractional bias, root mean square, normalized root mean square, and geometric variance and correlation functions. Further details of this analysis are discussed in Section 3.1.8.

2.14 LFG Collection and Utilization

It is important that the LFG generated at landfills should be collected and utilized to avoid environmental hazard, as well as, health risk. Besides the potential threats, the LFG collected could be beneficial and in the case of large landfills, an income generator. In most countries, regulatory requirements have urged the landfill owners to install a proper LFG control technology (ATSDR, 2001). Control technologies are often installed in landfills to collect, control and treat gases and use gases to benefit the community. There are two types of gas collection system: (1) the passive gas collection system; and (2) the active gas collection system. A passive gas collection system relies on pressure or concentration gradient to vent LFG into the atmosphere or a control system. Collection wells with vertical vents and gravel trenches are used to collect the LFG either during the active operation of landfill or after closure (ATSDR, 2001).

Whereas in an active system, it includes vacuums and pumps to move gas out of the landfill and piping that connects the collection wells to the vacuum. Vacuums or pumps pull gas from the landfill by creating low pressure within the collection wells. The active system basically consists of vertical gas wells and horizontal collectors. **Table 2.9** shows the comparison between the two collection systems.

Table 2.9: Comparison between passive and active LFG collection system (ATSDR, 2001).

	Passive	**Active**
Initial & operating cost	low	high
Income	low	high
Pressure gradient	positive	negative
Gas collection	inefficient	Efficient and functions with various gas system
Capacity	minimal	maximum

In a case study done in Thailand among three different landfill types namely semi-aerobic, anaerobic and gas to energy landfill, showed that the gas to energy landfill with active LFG collection systems had a higher operating, as well as, highest net present value (NPV, profit) (Pattharathanon *et al.*, 2012). The gas to energy landfill had a NPV of 50 Thai Baht for every tonnes of MSW in one day while the semi aerobic and anaerobic landfills had NPVs of 5-10 Thai Baht for every tonne of MSW per day (Pattharathanon *et al.*, 2012).

In the USA, LFG is the largest contributor to the man-made CH_4 emission contributing to almost 40% of the total CH_4 emission each year (USEPA, 2004). Several landfills in the USA have reported collecting about 100Nm^3 of CH_4 per ton of MSW landfilled in a given year (Themelis & Ulloa, 2007). Adding on to this, a total of 912,000kW or approximately 8 billion kWh/annum of electricity was generated by 295 LFG recovery facilities in the USA. Generally, the efficiency of LFG recovery system in a landfill is between 50-60% (Börjesson *et al*., 2007) but with the state of art CH_4 control system the LFG recoveries can rise to about 97% (Spokas *et al*., 2006).

Likewise, LFG could be used as an alternative fuel source. Shrestha *et al*., (2012) reported that LFG has about half the heating value at 16,785 – 20,495 kJ/m^3 (450 to 550 Btu/ft^3) as compared to 35,406 kJ/m^3 (950 Btu/ft^3) of natural gas. It is also a much cheaper fuel source than natural gas. Thus, LFG can be utilized as an alternative energy source which reduces the risk of explosion within a landfill, provides financial benefits for the community, conserves other energy resources and potentially reduces the risk of global climate change (ATSDR, 2001). **Figure 2.11** illustrates the potential usage of LFG collected from landfills as fuel for combustion as well as for power generation. Where as in smaller landfills where such utilization procedures are not applicable, the LFG is flared to become CO_2 to reduce the global warming input of CH_4 through direct release.

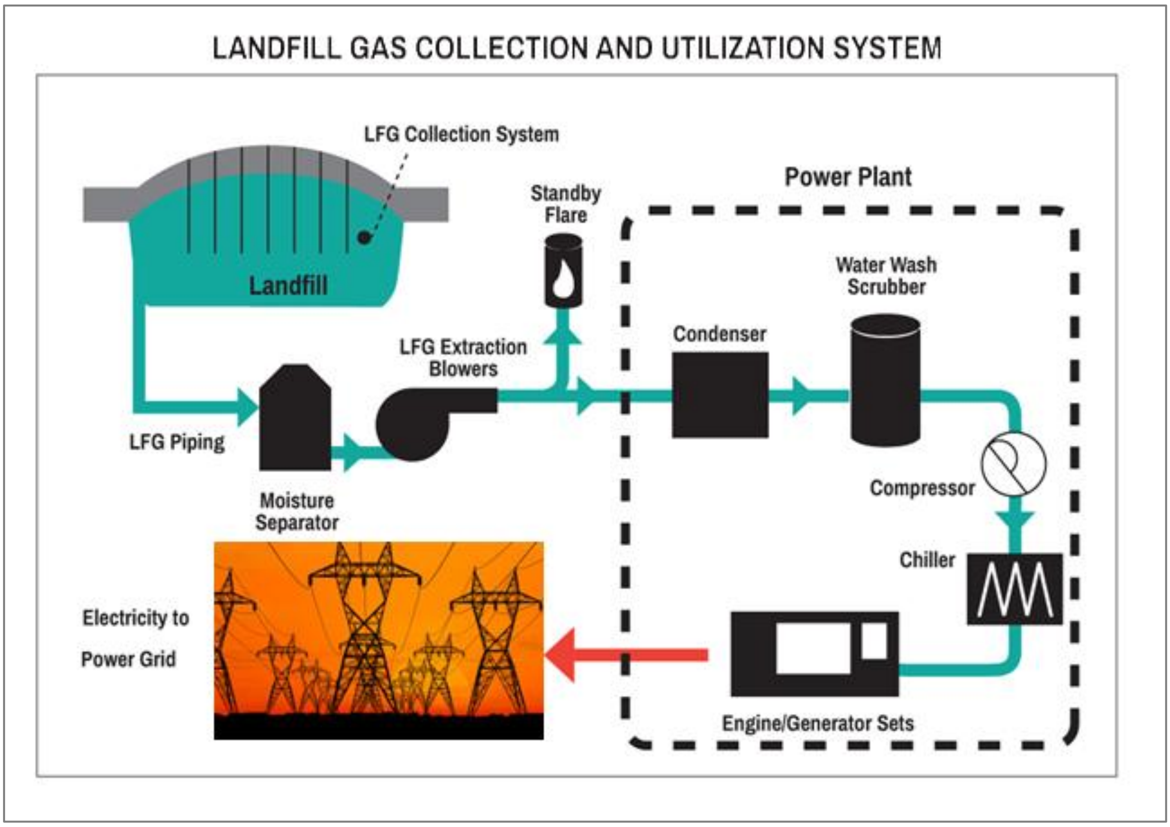

Figure 2.11: LFG collection and utilization system

2.15 Surface emission of landfill gas

The surface emission of LFG from landfill resources are through diffusion, advection via Darcy Flow and wind induced advection (Scheutz *et al.*, 2009). The diffusion mechanism is caused by LFG concentration variations in soil while advective transport is due to the pressure gradient. The pressure gradient between landfill and the atmosphere induces further LFG emission through advection. Thus, without proper LFG control technologies, the landfills are significant contributors to the atmospheric load of GHG. However, before any LFG control system is installed, it is important to quantify the amount of CH_4 that is being generated and emitted from the surface of the landfill into the atmosphere. Till-to-date, there have been numerous field studies undertaken worldwide to measure the CH_4 emissions from landfills through CH_4 fluxes. The fluxes are normally reported as CH_4 emission rate per unit area.

While, in a separate study by Bogner *et al.* (1997) the direct field measurements of landfill CH_4 emissions at small scale ($<1m^2$) varied over seven order of magnitudes (0.001 - >1000g CH_4/m^2/day). This difference in magnitude results from variation in waste composition, cover materials, soil moisture, temperature and other eminent variables (Bogner *et al.*, 2007). Therefore it can be said that the spatial variability of CH_4 emissions is higher than the temporal variability.

2.15.1 Methods used to measure surface emission of CH_4 from landfills

Taking into consideration the spatial and temporal variability of CH_4 surface emission from landfills, several methods to measure CH_4 fluxes from landfills have been developed, tested and adapted (Bogner *et al.*, 2007). However, there is no single method that is widely recognized or preferred to measure the annual average of CH_4 emissions (Oonk, 2010). Normally, the choice of measurement is dependent upon the purpose of the measurement, desired scale, cost and the available technology. The methods are divided into 3 categories: (1) above ground methods; (2) ground level methods; and (3) below ground methods. All these methods have their uniqueness, as well as, advantages and disadvantages. **Table 2.10** summarizes the types of measurement methods commonly used to measure the surface emission of CH_4 including its advantages and limitations.

Table 2.10: Types of measurement methods used to measure surface emission of CH_4 (Scheutz *et al.*, 2009)

Application	Status and references for method	Advantages	Disadvantages
Static chambers Single point measurements for chamber deployed at surface for variable time periods (usually less than 1 h). Measures diffusional flux based on change of gas concentration within chamber vs. time.	Most common technique for landfill emissions. Field validated for many terrestrial ecosystems and landfill settings. (Rolston & Klute, 1986)	A technique suitable for both positive and negative fluxes. Able to quantify very low fluxes. Simple to deploy with supporting analytical capability (Gas chromatography detector).	It is labour and time intensive. Appropriate geostatistical techniques are need for larger measurement area. Ay cause temporary disturbance of surface soil.
Dynamic chambers Single point measurements at ground surface using a chamber which continuously flushed with air or a flux gas. Measures flux based on the gas flow and difference between incoming and outgoing chamber CH_4 concentrations.	A common technique which has been field validated for terrestrial ecosystems and landfills.(Rolston & Klute, 1986)	Similar advantages as static chambers except it cannot measure the negative fluxes.	Same disadvantages as static chambers. Not able to quantify very low fluxes because of dilution of chamber gas with flux gas.
Soil core measurements Soil collected to determine bacterial activity. Soil sample is exposed to high concentration of CH_4 and decrease over time is measured. Also measures oxidation capacity of soil.	Measurements are done on ground surface. CH4 and CO2 concentration gradients in soil are indication of diffusion of gases through the layer. (Bogner *et.al*, 1995)	Gives the fundamental steps leading to emissions.	It has a very low spatial temporal resolution and does not take into account emissions caused by convection. Labor-intensive too.
Field flame ionization detector (FID) technique Measures the atmospheric CH_4 at a point directly above the landfill surface.	Widely used	Easy and simple to be practised	
Gradient techniques Measurement of below-ground gas concentration and pressure gradients with calculation of flux using equations for diffusional and pressure-driven flux.	Limited landfill field measurements but the basis for many transport models.(Glinski & Stepniewski, 1985)	It is used to measure the relative contribution of convective flux, diffusion and the influence of short term changes of barometric pressure, wind driven flux and passing storm fronts.	When there a cover soil and variable saturation in waste, the temporal points are often difficult to interpret.

Table 2.10 (Cont.)

Mass Balance method Measurements of CH_4 fluxes before and after the landfill is done using two imaginary vertical planes. CH_4 flux is measured as a product of wind velocity at each point.	Product of wind velocity and difference in CH4 concentration is used to calculate CH4 emissions (Oonk, 2010)	It is based on a simple and robust principle. It only needs wind direction and velocity as for emission calculation.	The instruments used are complicated, expensive and less flexible. CH_4 emission is underestimated at certain areas like slope.
Tracer techniques Rely on the concurrent measurement of atmospheric CH_4 and a tracer gas (usually SF_6 or N_2O). The tracer is released at a known rate; then the rate of CH_4 release can be determined from the ratio of CH_4 to tracer in the downwind plume.	A common technique which has been field validated for terrestrial ecosystems and landfills.(Czepiel *et al.*, 1996; Börjesson *et al,* 2007)	Takes into account the whole landfill including the edge leakages. It gives an integrated measurement of flux.	Not suitable for various topography at landfill and cannot quantify negative fluxes. Needs tracers gases such as SF_6 and N_2O which are potent GHG(s).
Micrometeorological techniques A group of techniques which measure the turbulent transfer of gases between the land surface and the lower part of the atmosphere. Rely on tower-mounted fast-response sensors to measure vertical gradients of wind speed and direction, CH_4 concentration, and other variables.	Widely used for landscape scale fluxes from terrestrial ecosystems. Has been sparingly used for landfill CH_4 fluxes. (Laurila *et al*., 2005)	Gives an integrated measure of landscape scale fluxes. Best used for flat terrain. It is useful for short-term and studies under changing atmospheric conditions.	Expensive technique which requires special equipment and cannot be applied at various topographies. Also not suitable for negative fluxes (only uptake of CH_4)
Remote sensing and other regional techniques Remote sensing techniques rely on tower- or aircraft-mounted sensors to measure atmospheric CH_4 and other variables. Flux is determined through models and/or scaling up of micrometeorological techniques. Regional techniques encompass both bottom-up and top-down techniques, including inverse modelling.	Used for regional or larger scale atmospheric studies, especially complex regional air quality studies examining spatial and temporal variations. Robust methods and models specific for landfill CH_4 not yet field-validated.(Modrak *et al*., 2007)		

2.15.2 The flux chamber measurement

The static and dynamic chamber measurement or also known as flux chamber measurement method were reviewed by numerous researchers through an extensive body of data gathering (Scheutz *et al.*, 2011, 2009; Wongyao *et al.*, 2010; Ishigaki *et al.*, 2008; Fourie & Morris, 2004; Spokas *et al.*, 2003). The operating principals of chambers are simple and flexible. The cost is low and it is also portable. It is generally operated by restricting the volume of air with which the gas exchange occurs to quantify changes in concentration of gases in the head space (Denmead, 2008).

In the static or closed chamber, the air circulation in and out of the chamber is hindered. Usually periodic air samples are taken from the head-space with a gas syringe and their concentration is measured using gas analyzers like gas chromatography. While in a dynamic or flow-through chamber, a constant flow of outside air is maintained through the headspace of the chamber and the difference in concentration between the air entering and leaving the head space is measured as flux (Denmead, 2008). In this study, the closed flux chamber made of plexi glass as discussed in Section 3.9.1 was used.

In Europe, the USA and South Africa, the flux chamber measurements were the predominant method for quantifying surface emission of landfill CH_4 (Scheutz *et al.*, 2011; Galle *et al.*, 2001; Börjesson *et al.*, 2000; Börjesson, 1997; Czepiel *et al.*, 1996). This method precluded to be inexpensive while still providing meaningful results.

Disadvantages to this method are normally spatial limitations as well as labor constrains.

In a study by Tregoures *et al.,* (1999), seven methods to measure CH_4 flux from landfills were evaluated at French landfill sites with regard to scientific, technical and economic criteria. The study found that the chamber methods provided the best overall measurement strategy in terms of accuracy, flexibility and robustness. This technique is also commonly used for CH_4 fluxes measurements both before and after placement of landfill cover layer (Fourie & Morris, 2004). However chambers with small chamber volume and short gas extracting time (<3 minute), showed inconsistent CH_4 flux measurement compared to other indirect methods of measurements (Börjesson *et al.*, 2000).

Hence, suitable chamber volume is required to obtain a representative result. Livingston & Hutchinson (1995) concluded that flux measurements on sites with large exchange rate are best served by chambers with large volume to area ratios (V:A) typically greater than 150 Lm^{-2}. Other researchers have scaled the range of ratio between 60 Lm^{-2} to 300 Lm^{-2} and with measuring period ranging from 20 to 40 minute (Abushammala *et al.*, 2012; Senevirathna *et al.*, 2007; Denmead, 2008;Bogner *et al.*, 1997; Reinhart *et al.*, 1992;).

Chambers are normally made out of stainless steel, poly-vinyl chloride (PVC) and fibre-glass. The chamber used in this study was designed according to the specifications above. Details on the design and volume to area ratio are explained further in Section 3.9.1.

2.16 Meteorological factors influencing the surface emission of LFG

The importance of the meteorological factors influencing the surface emission of landfill CH_4 has been observed on site by various researchers (Wang *et al.*, 2012; Czepiel *et al.*, 2003; Christophersen *et al.*, 2001; Galle *et al.*, 2001). In addition to this, gas flux models by Young (1990,1992), Naster *et. al.* (2001) and Poulsen *et. al.* (2003) have used barometric pressure as its key variable.

Figure 2.12 shows a conceptual diagram for LFG flow rate and composition, as well as, the connection between atmospheric pressure, temperature and other weather parameters. As a result, the LFG emission is delicately dependent on the atmospheric pressure variation which is induced by temperature, air intrusion and other meteorological factors (Gebert & Groengroeft, 2006). The atmospheric pressure perturbs the LFG pressure gradient in the landfill. Therefore, when there is positive LFG pressure gradient, CH_4 gas emission increases. However when a negative LFG pressure gradient occurs, the air is in-fluxed. The air influx reduces CH_4 to the atmosphere but increase the potential risk of explosion within a landfill.

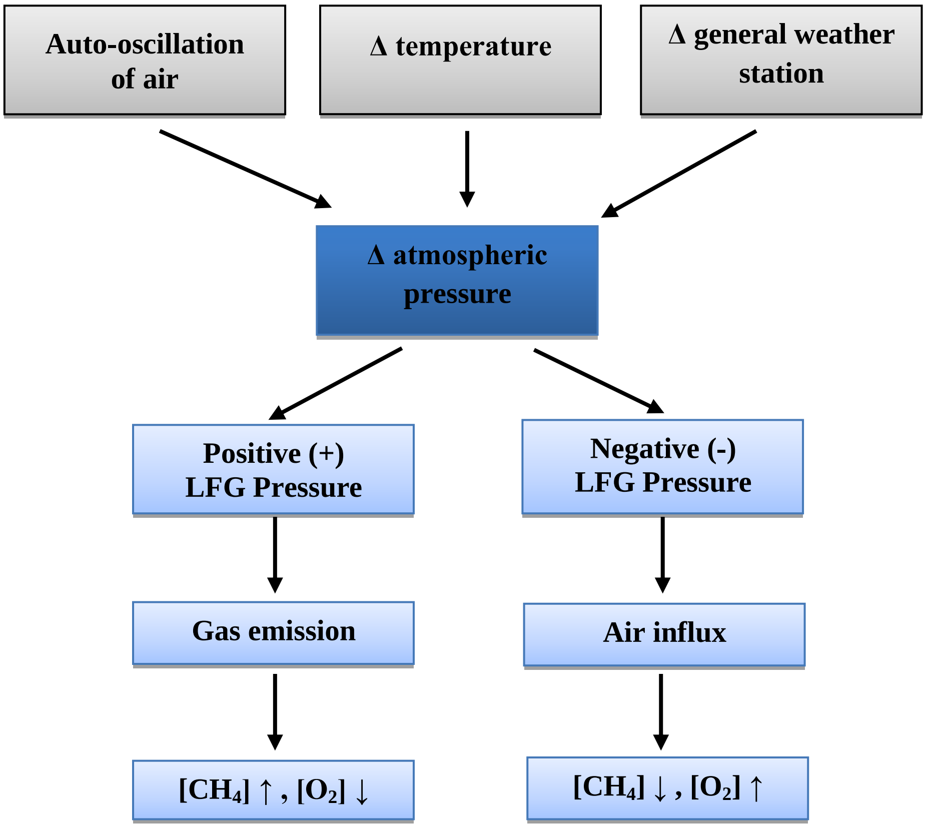

Figure 2.12: Conceptual diagram on the regulation of LFG emission in a landfill, Δ = difference, LFG = landfill gas, [CH$_4$]= methane concentration, [O$_2$] = oxygen concentration, ↑ = increase, ↓ = decrease, (Gebert & Groengroeft, 2006)

2.16.1 Atmospheric pressure

Czepiel *et al.*, (2003) examined the influence of atmospheric pressure on landfill methane emission from 12 individual emission tests and concluded that the measured emissions were negatively correlated with surface atmospheric pressure. The results from the test had a 0.93 correlation coefficient when modelled by linear regression. **Figure 2.13** shows the CH$_4$ emission and pressure relationship from the test (Czepiel *et* *al.,* 2003). The different shapes in the figure represents different period of measurement while the error bars indicate the coefficient of variation of each measurement.

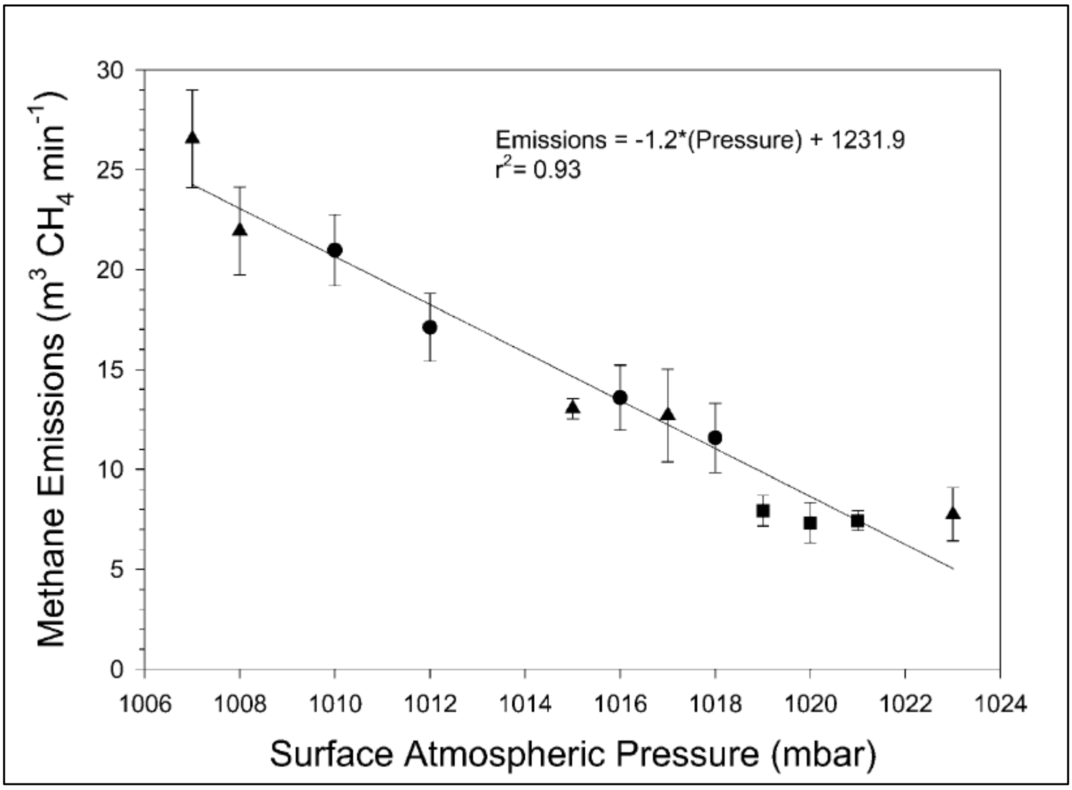

Figure 2.13 : CH_4 emissions vs atmospheric pressure. Solid squares are data from August 1996, solid circles are data from February 1997, and solid triangles are data from March and April 1997. The error bars represent the coefficient of variation of each measurement. The solid line is the linear regressions of these data with its equation of the regression line and r^2. Source: Czepiel *et al.*, (2003).

2.16.2 Rainfall

The effect of rainfall on the surface emission of CH_4 at landfills had contradicting evidences. In a study at nine solid waste disposal sites in Thailand, it was found that CH_4 emissions during the rainy season were about five to six times higher in magnitude compared to winter and summer season (Wangyao *et al.*, 2010). Comparatively in a research conducted at the Sudokwon Landfill in South Korea, a different reasoning was given to the higher CH_4 emissions during the rainy season (Park & Shin, 2001). Due to much ground water content during the rainy season, the surface pores at the landfill are blocked. Thus this accelerated the CH_4 flux through the remaining open pores. However when it was a sunny day, CH_4 is easily released through many pores on the surface and eventually escapes to the atmosphere (Park & Shin, 2001). So, quantitatively more CH_4 is emitted through the surface during the hot/sunny days.

2. 16. 3 Temperature

In general, the thermal regime of landfills is controlled by climatic and operational conditions of a landfill. The temperature in the landfill plays a key role in determining the long-term potential of LFG production and emissions. Wang *et al.*, (2012) concluded that high temperature (30˚C to 40˚C) accelerated the waste degradation and gas generation, thus increasing the higher possibility of surface emissions of CH_4. In an earlier study by Park & Shin (2001), they had examined the diurnal and seasonal variation of CH_4 from landfills. Results from the diurnal variation concluded that the surface CH_4 emission was scarce during the minimum temperature of the day and rose to a peak when the temperature was at a maximum. **Figure 2.14** shows the variation of flux rate according to time of the day. A significant peak of LFG was generally obtained during the hottest time of the day which is around 2pm (Park & Shin, 2001). Viewing the recorded of the seasonal variation, they summarized that the CH_4 flux rate decreased from summer to winter.

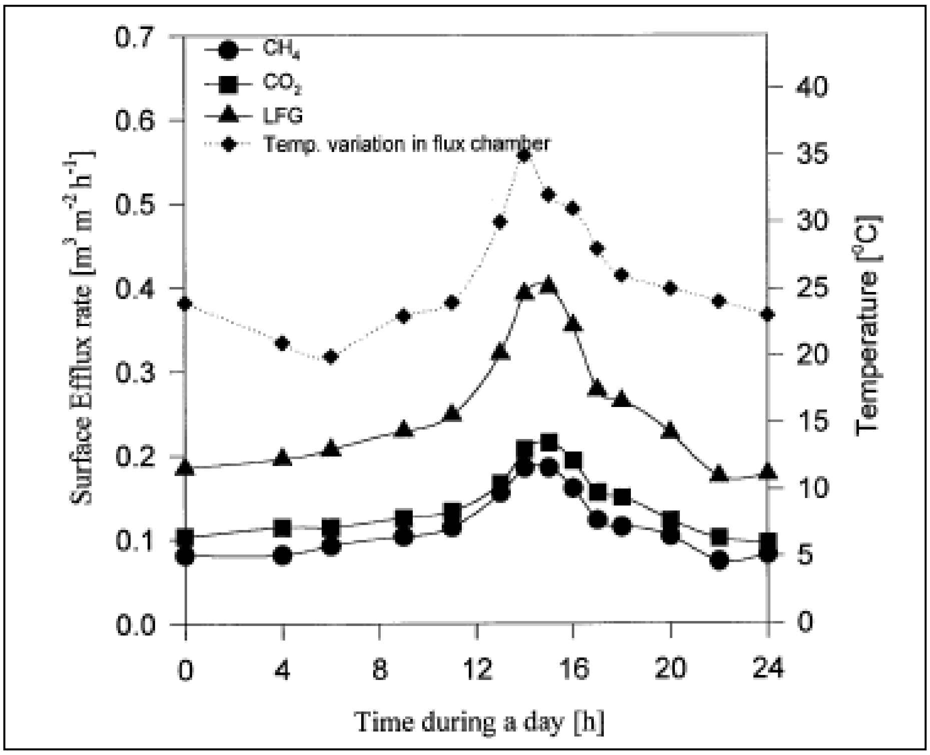

Figure 2.14: Variation of LFG flux according to the time of the day *(Park & Shin, 2001)*

The influence of ambient temperature on the LFG emission was studied by researchers in the past (James *et al.*, 2008; Yesiller *et al.*, 2008; Einola *et al.*, 2007; Townsend *et al.*, 1996; Attal *et al.*, 1992) and was mostly concentrated in temperate countries like Alaska, British Columbia, Michigan, Finland and New Mexico. Alternatively, Robinson (2007) found that the rapid transition from acetogenic to the methanogenic phase of the landfills in the warmer climates were two to three times faster than that of the landfills in temperate countries. Blakey *et al.*, (1997) had summarized that temperature is an important factor which affects the CH_4 content in LFG. So it is important for landfills to operate under optimum temperature for faster LFG production and refuse stabilization (Wang *e. al.*, 2012).

2.17 Landfill cover

The two main recommendations put forward by the IPCC as strategies for potential mitigation of CH_4 from the waste sector are (1) the enhancement of landfill CH_4 recovery and utilization; and (2) the optimization of methanotrophic CH_4 oxidation in landfill cover soils and biofilter (Polettini, 2012; Huber-Humer, 2011; IPCC, 2007). The former strategy are often adapted in most of the larger landfills while the latter strategy is a common practice in smaller and older landfills which do not produce much LFG for gas recovery and utilization (Chanton *et al.*, 2011; Stern *et al*, 2007). However, a landfill cover is an essential mitigation practice in both larger and small landfills as an effort to reduce lateral migration of CH_4 from the landfill cover. Therefore, the landfill cover soil acts as the environmental interface between the waste layer and atmosphere (Jing *et al.*, 2011). Landfill cover with the potential microbial activities from methanotrophs has been frequently verified in the past years (Huber-Humer *et al*., 2008; Scheutz *et al*., 2009) and the following benefits were identified:

a) reduction of gas emissions especially CH_4
b) protection of the sealing element (e.g. capillary barrier)
c) growth of vegetation; ability of root penetration and sustainable nutrient supply
d) optimization of water balance and water holding capacity
e) prevention of direct contact between harmful substances and environment
f) stability especially at long and steep slopes

2.17.1 Different types of cover material

In a review by Scheutz *et al.*, (2009), different types of cover materials had been used as the landfill cover to obtain maximum CH_4 oxidation capacity to reduce the impact of CH_4 emission to the environment and health. Each of these materials was studied in the laboratory using the incubation experiments (batch test) or in continuous gas flow system (packed column test) to simulate and control the external factors involved in the actual environment. Among the materials used were sand, coarse sand, humid soil, clay, landfill soil and waste soil from landfill reactor. The materials tested were later applied to field studies to evaluate its actual performance in CH_4 oxidation. It was noted that in both tests, the CH_4 oxidation rate in landfill cover soil has been more than 100 µg CH_4 g^{-1} h^{-1} and 200 g CH_4 m^{-2} day^{-1}, respectively (Scheutz *et al.*, 2009).

2.17.2 Biocover

In the 2007 IPCC AR4 report on climate change mitigation efforts, biocovers and biofilters were listed as the key mitigation technologies and practices to reduce GHG emissions from landfills (Bogner *et al.*, 2007). Biocovers are organic filter material which supports the growth of methanotrophs for CH_4 oxidation when placed upon a gas distribution layer made of gravel (Pedersen *et al.*, 2011).

Biocovers are normally rich in organic matter and from previous studies it is evident that this condition enhanced the microbial CH_4 oxidation in landfill cover soil compared to the use of pure clay cover (He & Ruan, 2008; Stern *et al.*, 2007; Abichou *et al.*, 2006). He *et al.*, (2008) established that, compared to a clay soil; biocovers made from organic waste products like compost could reduce CH_4 emission to the atmosphere by about 37.4 kg CH_4 per m^2, annually. The biocover is routinely spread over a certain level of depth above the waste layer to act as a medium for CH_4 oxidation.

2.18 Methane oxidation in landfill cover

Scheutz *et al.,* (2009) reported that around 20cm to 40cm below the surface, in zones where vertical profiles of CH_4 and O_2 overlap, the CH_4 oxidation is at its greatest. CH_4 oxidation occurs at the interface of aerobic and anaerobic zones, where CH_4 is generated in anaerobic regions below, and methane uptake occurs in aerobic zones above. The first experiment to quantify methane oxidation in landfill cover was done by Whalen *et al*., (1990) using laboratory batch experiments. 45g CH_4/m^2d (= 63L CH_4/m^2d) was rapidly oxidized in a landfill topsoil in a closed municipal solid waste landfill where the CH_4 concentration was between 1 ppm to 10,000 ppm. Capacity for CH_4 oxidation was observed in landfill cover soil in column experiments, with rates as high as >200 g CH_4/m^2d (Scheutz *et al*., 2003; DeVisscher *et al*., 1999). High methane oxidation capacity could be found in porous, coarse and organic-rich substrates (Croft and Emberton, 1989).

2.19 Methanotrophic activity in landfill cover

Methanotrophs are a physical group of bacteria that are unique in their ability to utilize CH_4 as carbon and energy source. Whalen *et al.* (1990) was the first to recognize the importance of methanotrophs in landfill soils followed by a more comprehensive review on the properties of methanotrophs by Hanson & Hanson (1996). The former had estimated that methanotrophs were able to convert about 50% of the CH_4 emitted from the landfill surface while the latter was able to define the characteristics of the methanotrophs and narrate the complete pathway for the microbial oxidation of CH_4 to CO_2. **Figure 2.15** shows the pathway for the microbial oxidation including the intermediate steps of CH_4 oxidation into methanol, then methanol into formaldehyde (CHOH), the subsequent oxidation of formaldehyde to formate (CHOOH) and finally into CO_2.

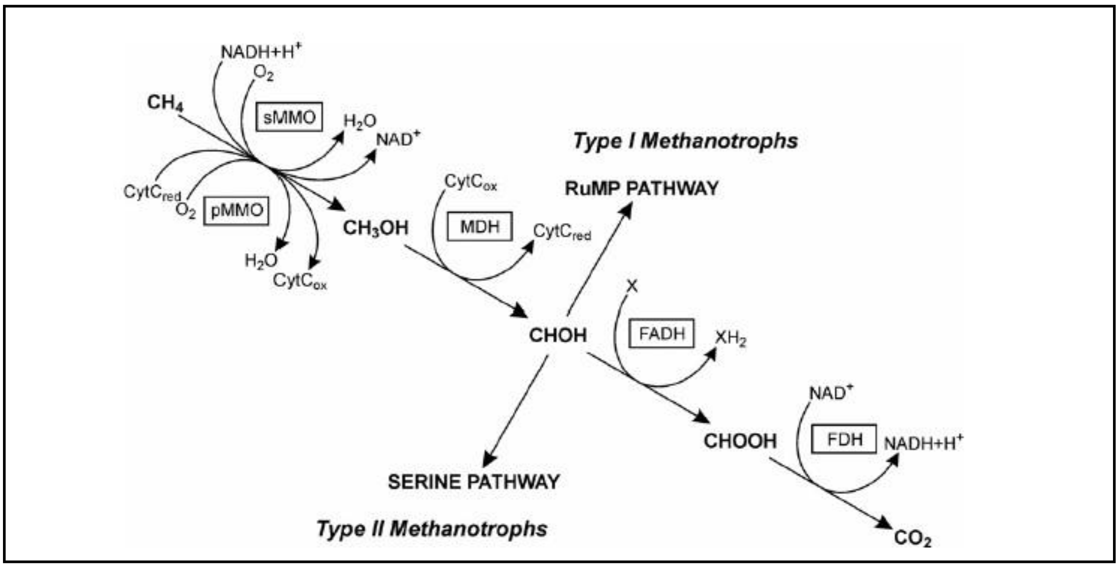

Figure 2.15: The pathway for methane oxidation and assimilation of formaldehyde by Methanotrophs (Hanson & Hanson; 1996 and Scheutz *et al,;*2009).

2.20 Factors influencing methanotrophic activity in landfill cover

There are a number of factors that influenced the methanotrophic activity in the landfill cover. The most important factors are temperature, moisture content, pH, CH_4

concentration, O_2 supply, organic content and nutrients (He & Ruan, 2008; Einola *et al.*, 2007; Hanson & Hanson, 1996;). Thus, these parameters are very important in controlling CH_4 oxidation in a landfill. Hanson & Hanson (1996), have also observed that a shift in any of these environmental stimuli response to changes in the methanotroph population in the cover soil.

2.20.1 Temperature

Temperature is one of the most important element for the methanotropic activity in the cover soil. The optimum temperature helps to maximize CH_4 oxidation with addition to construct a better landfill cover. Since most methanotrophs are mesophiles, the optimum temperature range for CH_4 oxidation in soil environment was around 25˚C to 35˚C (Scheutz *et al.*, 2009; Hanson & Hanson, 1996). Einola *et al.*, (2007) also reported similar optimum temperature which ranged from 20˚C to 38˚C. While in temperate countries during winter when the atmospheric temperature is between 5˚C to 10˚C, CH_4 oxidation is reduced significantly or even reaches a plateau. The Q_{10} describes the value for number of times the oxidation rate increases when temperature is increased by 10˚C at temperature below the optimum temperature. Higher Q_{10} value indicates higher CH_4 oxidation. For temperatures ranging from 10˚C to 30˚C, the Q_{10} value was observed exponentially between 1.7 to 4.1 suggesting that mesophilic methanotrophs are the main consumer of CH_4 in the biocover soil (Scheutz *et al.*, 2009; Park *et al.*, 2005; Scheutz & Kjeldsen, 2004; Czepiel *et al.*, 1996).

2.20.2 Moisture Content

The moisture content typically follows a parabolic curve on the CH_4 oxidation rate in the cover soil. This indicates that the CH_4 oxidation is reduced at both very low and high moisture content of the cover (Scheutz *et. al*., 2009; Einola *et. al.,* 2007; Park *et. al*., 2005; Scheutz & Kjeldsen, 2004). Since moisture content within the soil is the main transport medium of the nutrients for microbial activity, it is an essential factor for CH_4 oxidation. Besides microbial activity, the optimum soil moisture content is also important to maximize gas phase molecular diffusion of LFG. In addition to this, soil texture, pore size distribution and water retention capacity of the soil also aide the gaseous transport. From previous literatures, the optimum soil moisture content for landfill cover soils ranged between 10% to 20% of wet weight basis and 45% to 110% of dry weight basis (Scheutz *et al.*, 2009; Mor *et al.*, 2006; Czepiel *et al.*, 1996; Whalen *et al.*, 1990). The moisture content of the cover soil was closely related to the temperature range of the cover soil (Einola *et al.*, 2007).

2.20.3 pH

Previous studies suggest that the optimum pH for CH_4 in the cover soil was between pH 5.5 and pH 7.5 (Scheutz *et al.*, 2009; Scheutz & Kjeldsen, 2004). This pH range maximizes the growth of methanotrophs and is in consistent with the pure cultures of methanotrophs by Hanson & Hanson (1996). Similar results of pH range were also presented by Navarani (2009), Sitiaishah (2011) and Jayanthi (2013) for maximum CH_4 oxidation. According to Pawloska (1999), no significant methanotropic activity was observed between pH 7.61 and pH 8.89. The pH of the cover material used in the studies were lowered using acid substances and raised using lime for studying the optimum pH for methanotrophic activities for maximum CH_4 oxidation. The pH used was from pH 5 to pH 8.

2.20.4 Methane concentration

The response of CH_4 oxidation within cover material or soil varies with the CH_4 concentration. The vertical profile of the cover material is exposed to different amount of CH_4. The deepest layer of the landfill cover will be exposed to the highest amount of CH_4 (He & Ruan, 2008). Previous studies by Jayanthi (2013), Navarani (2009) and Perdikea *et. al.* (2008), showed that cover material between the 40 to 60 cm thickness is sufficient for CH_4 oxidation. Therefore, it is essential for the cover material to be implanted to a certain depth to produce maximum CH_4 oxidation. Cover material which are too thick or too thin are not cost effective and a waste of resources. It is important to the CH_4 content in the LFG for effective cover material to be applied. Maximum CH_4 oxidation is said to occur when suitable environment exists with the presence of methanotrophic bacteria, optimum O_2 and CH_4 ratio and good retention time (Jayanthi, 2013).

2.20.5 Oxygen supply

Oxygen is required to oxidize CH_4 produced by the methanotrophs. The stoichiometry equation below states that two molecules of O_2 per molecule of CH_4 are needed for microbial CH_4 oxidation as shown below:

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O + \Delta\, energy\ (210.8\ kcal\ mol^{-1})$$

However in most of the landfills, O_2 penetration depth will often be the limiting factor of CH_4 oxidation process. In relation to this, soil texture and composition, pore size and porosity are important parameter to aide this process. However, O_2 supply into the cover soil also deprived from CH_4 when landfills use low permeable cover to hinder rainfall infiltration and subsequent leachate production (Scheutz *et al.*, 2009). In a study by Wilshusen *et. al.* (2004), it was established that an O_2 range between 0.45 to 20% supported maximum CH_4 oxidation.

2.20.6 Organic content and nutrient

Huber-Humer *et al*., (2009) recommended the minimum organic content in any biocover or cover material to be 15% of its dried mass or weight. This value was used as a threshold criterion to access the performance of different biocover or cover material used in CH_4 oxidation (Pedersen *et al.*, 2011; Huber-Humer *et al*., 2009). Another important factor is the availability of nutrients such as N in the cover material. N, specifically inorganic N in the form of ammonia ($NH4^+$) and nitrate (NO_3^-) helps to stimulate or inhibit CH_4 while oxidizing it in the cover material. (Scheutz *et al*., 2009).

2.21 Organic waste as Biocover

Many researches have used various materials as biocover for CH_4 oxidation in landfills. Sand, clay and soil are among the commonly used materials as it is easily available to the landfill operators. Although these materials have been good landfill caps but CH_4

oxidation rate remains low in some cases. This result emphasized the importance of texture, pore size and porosity of the cover material for the enhancement of microbial CH_4 oxidation (Rachor *et al.*, 2011; Maurice & Lagerkvist, 2004).

The use of organic waste such as compost as a biocover material has been given due importance for its high CH_4 oxidation rate in a landfill cover. Among the organic waste commonly use a direct or supplementary material for landfill covers are food waste, agricultural waste, garden waste, animal manure and sewage sludge. This sort of manipulation of landfill covers or better known as engineered materials provides a promisingly complementary strategy for the control of CH_4 emission.

2.21.1 Compost

Compost is mixture of organic residue and soil or organic residue only that has been piled, moistened and have undergone aerobic biological decomposition. Compost is normally made from residues of garden waste, agricultural waste and animal manure. Compost is normally rich in organic matter and a suitable medium for methanotroph activities. A number of studies by Abichou *et al.,* (2006), Stern *et al.,* (2007) and He *et al.* (2008) also concluded that compost rich in organic matter showed better microbial CH_4 oxidation compared to pure clay cover. While in year 1999 and 2001, researches done by Humer & Lechner (who studied compost made from MSW and sewage sludge as a cover material) discovered that 100% CH_4 oxidation was possible with the use of these materials. Furthermore, engineered biocovers like this have been known to effectively perform multiple annual cycles of CH_4 oxidation in northern temperate climates (Huber-Humer, 2004).

In separate studies performed by Navarani (2009), Sitiaishah (2011) and Jayanthi (2013), the performance of compost made from grass clippings and cow manure were examined in laboratory scale batch and packed column test for CH_4 oxidation rate. All compost used showed 100% CH_4 oxidation capacity varying only in the number of days for the maximum CH_4 oxidation. Therefore, this proves that compost is an effective cover material for CH_4 oxidation.

2.22 Response Surface Modeling

Response Surface Modelling (RSM) is used to design the best combination of experiments needed to choose the optimum parameters without carrying out large number of experiments to save time and resources (Guo *et al.*, 2009; Bhunia & Ghangrekar, 2008). This method which uses the mathematical modelling and optimization studies using a three-factor, three-level Box-Behnken experimental design for excellent predictability and fewer experiments compared to other designs (Yetilmezsoy *et al.*,2009). In this study, the experimental design was applied to determine optimum pH, moisture content and temperature to determine the optimum parameter required for maximum CH_4 oxidation within the biocover material.