

## Chapter 1

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# Hazardous Emissions from Municipal Solid Waste Landfills

### 1. Introduction

Last decades continuing industrial and commercial growth in many countries around the world has been accompanied by rapid increases in both the municipal and industrial solid waste production. During the latter part of the 1990s, annual waste production ranged from 300 to 800 kg per person in the more developed countries to less than 200 kg in other countries. In 2009, Polish population produced 12 million Mg of MSW, but only 9,3 million Mg had been collected, namely 246 kg per person. The sanitary landfill method for the ultimate disposal of solid waste material continues to be widely accepted and used due to its economic advantages. Comparative studies of the various possible means of eliminating solid municipal waste (landfilling, incineration, composting, etc.) have shown that the cheapest, in term of exploitation and capital costs, is landfilling.

The EU Landfill Directive introduces specific targets for waste disposal reduction and very stringent technical requirements for landfilling and landfill facilities. Despite this directive, the situation is not homogeneous across Europe. Eurostat's data for the year 2010 shows, that several countries are very advanced in diverting municipal waste from landfills, often due to the implementation of national measures to reduce landfilling of municipal waste. Switzerland, Germany, the Netherlands, Sweden, Austria, Denmark and Belgium have reported landfill rates below 5%. In the new Member States and the Candidate Countries as well as in Iceland, landfilling is still the predominant waste management option. Landfill rates in these countries range between 62% in Slovenia and 100% in Bulgaria. In 2009, 85% of collected municipal solid waste in Poland was landfilled.

As it was mentioned earlier, for technical, economical and regulatory reasons, landfilling in most countries remains as the most practical waste treatment solution. For many other reasons, it also appears the least rational approach to waste

management. Land repositories become committed to waste disposal for perhaps 10 years and the aftercare period may last up to 100 years. Biogas and leachate may have severe impacts on the environment, and landfilling may cause other types of public nuisance. Furthermore, in many countries municipal solid waste (MSW) is deposited to landfills with no waste separation and no proof of contents. Even household waste contains a component of hazardous substances that is potentially harmful, amounting to 1% of MSW. Major developments have now occurred with respect to waste management and in its legislative control, such that recycling targets, and biodegradable organics pretreatment prior to landfilling are now limiting the rate of landfill expansion.

Wastes landfilling causes two main types of pollution, which correspond to the migration into the natural environment of:

- leachates, defined as water that has percolated through the wastes (rainwater or groundwater seepage), a source of soil and groundwater contamination, and
- biogas produced by the fermentation of organic matter, a source of air pollution.

Some alternative methods such as recycling, composting and incineration are nowadays very much encouraged but even incinerations create residue of approximately 10-20% that must be ultimately landfilled. At present, modern landfills are highly engineered facilities designed to eliminate or minimize the adverse impact of the waste on the surrounding environment.

In this chapter the most common landfill impacts on environment such biogas, and landfill leachate emission, landfill fires, and its' contribution in human health threat have been presented and characterized. New waste management strategies, including sustainable landfilling, also have been reported, as an example of reducing of environmental impacts of waste landfilling.

## **2. Landfill as a source of pollution**

The main emissions (leachate and biogas) are greatly influenced by biological processes taking place in the landfill. When MSW is landfilled without pre-treatment, emissions arise during landfill operations and continue after closure. Depending on waste composition, climatic conditions, etc., these constitute approximately 150 (ranging from 70 to 300) m<sup>3</sup> biogas/Mg MSW (based on dry weight) and about 5 m<sup>3</sup>·ha<sup>-1</sup>·d<sup>-1</sup> of highly polluted leachate, as an approximation for a high compacted landfill in Central Europe at an annual precipitation rate of 600-750 mm.

The biogas produced must be extracted and flared or can be used as an energy source. The leachate produced has to be collected and treated.

### **2.1. Biogas**

One of the products of biological processes running inside the MSW landfill body is biogas. It is an effect of biodegradable organic matter decomposition,

and the rate of biogas generation and its composition changes during landfill life cycle (Fig. 1). It is based on a combined model of the bacterial decomposition of waste (Phases I–IV) and the model for the long-term behaviour of landfill gas from old waste deposits (Farquhar 1973). The first three Phases (I–III) of the landfill gas production of non pre-treated waste last for a half up to three years. In Phase I, the aerobic phase, aerobic bacteria decompose the long molecular chains of complex carbohydrates, proteins and lipids by oxidising them into carbon dioxide. The emitted gas consists mainly of nitrogen and oxygen. The amount of carbon dioxide increases as the conditions for the micro organisms improve. Phase II is the anaerobic acidic phase in which the oxygen in the landfill has been used up and anaerobic bacteria convert compounds produced in Phase I into acetic, lactic and formic acids and alcohols. The landfill becomes highly acidic. The emitted landfill gas consists mainly of hydrogen and carbon dioxide. The remaining nitrogen also is emitted into the air. Phase II is highly unstable and the reactions will stop if oxygen contacts the waste. During Phase III, the acids from Phase II are decomposed into acetate. Thus, the acidity of the landfill decreases. Methane-producing bacteria establish in the landfill body. Therefore, the production of methane increases and reaches a level of about 70%. The carbon dioxide production decreases from 70% to 40%. The landfill gas production in this phase is large. After the short Phases I–III, follows the long Phase IV, which could last for more than twenty years. In this phase, the gas production is relatively constant. The methane–carbon dioxide relation is about 1.25:1. The landfill gas production decreases in comparison to Phase III, but is still high enough to prevent ambient atmosphere to infiltrate the landfill body.

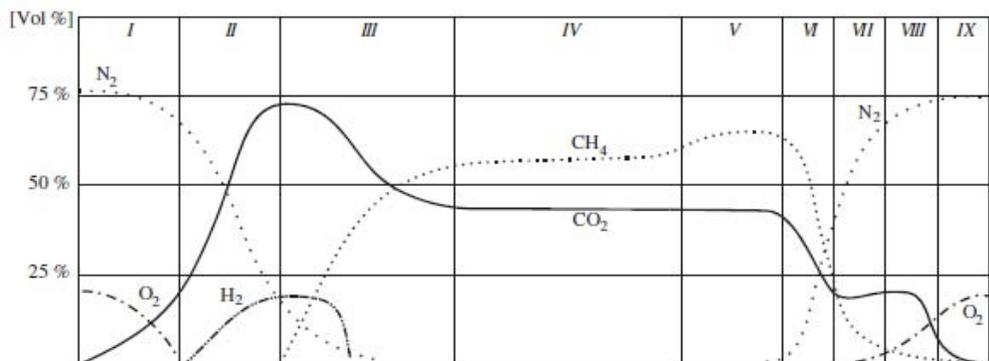


Fig. 1. Typical landfill gas production phases (Bockreis, Steinberg 2005, modified)

Phase V is the long-term phase in which most of the landfill gas emissions occur. The amount of carbon dioxide emissions decreases in relation to the methane emissions. The ratio of methane to carbon dioxide can increase up to values of about four. As in Phase V, the pressure inside the landfill body is higher than the pressure of the ambient atmosphere. Therefore, no fresh air comes

in contact with the waste. The air infiltration phase, Phase VI, is characterized by a decrease of the pressure inside the landfill body. Therefore, air enters the landfill body and the bacteria are supplied with oxygen, which results in a change of the composition of the emitted gas. In the upper tiers of the landfill body, the anaerobic processes stop and aerobic processes start. Thus the methane-carbon dioxide ratio drops to 0.6:1. This phase, as well as all the following phases, does not start with a homogeneous distribution over the landfill body.

Caused by the reduced gas production in Phase VI, more ambient air infiltrates the landfill body and due to the supply of oxygen the emitted methane is oxidized inside the landfill body in Phase VII. Emissions into the atmosphere are below detection.

In the following phases, the carbon dioxide phase (Phase VIII) and the air phase (Phase IX), the methane production nears zero and the carbon dioxide emissions decrease to a level of 5–20% in Phase VIII and below 5% in Phase IX. The concentration of nitrogen is about 78%, the oxygen emissions vary from 2% to 17% according to the carbon dioxide concentration (Farquhar, Rovers 1973, Bockreis, Steinberg 2005).

Biogas contains also other gases in traces, primarily sulfur compounds and volatile organic compounds (VOCs). The methane and the carbon dioxide are greenhouse gases like some trace gases in landfill gas, as for example the chlorinated and fluorinated organic compounds. Landfill gas has other effects on the environment: in addition to the explosive potential of methane, important risks and harmful effects to the environment and public health exist because of the presence of hydrogen sulfide ( $H_2S$ ), siloxanes, and of potentially toxic VOCs (benzene, vinyl chloride, dichloromethane, chloroform, toluene, dichlorobenzene, etc) as well as compounds responsible for odours (VOCs, sulfur compounds, etc).

More than 500 trace compounds in landfill gas have been identified. Concentrations of these trace components are very changeable. United States Environmental Protection Agency (US EPA) proposed to use LandGem software tool for landfill gas emission estimation. LandGEM uses the first-order decomposition rate equation to estimate annual emissions over a specified time period. One of the benefits of using LandGem model is the estimation of landfill gas potential, methane content and its calorific value. Another advantage is the possibility of assessment of trace pollutants emission. The typical concentrations of most common trace components of landfill gas has been proposed to be constant in this model, and has been shown in Table 1. Most of these compounds are toxic.

It is common understanding now that landfill gas (LFG) should be considered either as a significant source of pollution and risk (if migrating uncontrollably to the air and ground), or as a significant source of renewable energy (if extracted and processed accordingly). There are two possible solutions for dealing with LFG emissions. In case of low methane ratios, LFG should be extracted and flared or oxidized in biofilters.

Table 1

Trace components of landfill gas, and its' concentrations and harmful potential considered in LandGem model for pollutants emission into the air

Pollutant in landfill gas	Assumed concentration in landfill gas emission model – LandGem [ppmv]	Note
1,1,1-Trichloroethane (methyl chloroform) - HAP	0.48	A
1,1,2,2-Tetrachloroethane - HAP/VOC	1.1	A, B
1,1-Dichloroethane (ethylidene dichloride) - HAP/VOC	2.4	A, B
1,1-Dichloroethene (vinylidene chloride) - HAP/VOC	0.20	A, B
1,2-Dichloroethane (ethylene dichloride) - HAP/VOC	0.41	A, B
1,2-Dichloropropane (propylene dichloride) - HAP/VOC	0.18	A, B
2-Propanol (isopropyl alcohol) - VOC	50	B
Acetone	7.0	
Acrylonitrile - HAP/VOC	6.3	A, B
Benzene - No or Unknown Co-disposal - HAP/VOC	1.9	A, B
Benzene - Co-disposal - HAP/VOC	11	A, B
Bromodichloromethane - VOC	3.1	B
Butane - VOC	5.0	B
Carbon disulfide - HAP/VOC	0.58	A, B
Carbon monoxide	140	A, B
Carbon tetrachloride - HAP/VOC	4.0E <sup>-03</sup>	A, B
Carbonyl sulfide - HAP/VOC	0.49	A, B
Chlorobenzene - HAP/VOC	0.25	A, B
Chlorodifluoromethane	1.3	
Chloroethane (ethyl chloride) - HAP/VOC	1.3	A, B
Chloroform - HAP/VOC	0.03	A, B
Chloromethane - VOC	1.2	B
Dichlorobenzene - (HAP for para isomer/VOC)	0.21	B
Dichlorodifluoromethane	16	
Dichlorofluoromethane - VOC	2.6	B
Dichloromethane (methylene chloride) - HAP	14	A
Dimethyl sulfide (methyl sulfide) - VOC	7.8	B
Ethane	890	
Ethanol - VOC	27	B
Ethyl mercaptan (ethanethiol) - VOC	2.3	B
Ethylbenzene - HAP/VOC	4.6	A, B
Ethylene dibromide - HAP/VOC	1.0E <sup>-03</sup>	A, B
Fluorotrichloromethane - VOC	0.76	B
Hexane - HAP/VOC	6.6	A, B
Hydrogen sulfide	36	
Mercury (total) - HAP	2.9E-04	A
Methyl ethyl ketone - HAP/VOC	7.1	A, B
Methyl isobutyl ketone - HAP/VOC	1.9	A, B
Methyl mercaptan - VOC	2.5	B
Pentane - VOC	3.3	B
Perchloroethylene (tetrachloroethylene) - HAP	3.7	A

Table 1 continued

Pollutant in landfill gas	Assumed concentration in landfill gas emission model – LandGem [ppmv]	Note
Propane - VOC	11	B
t-1,2-Dichloroethene - VOC	2.8	B
Toluene - No or Unknown Co-disposal - HAP/VOC	39	A, B
Toluene - Co-disposal - HAP/VOC	170	A, B
Trichloroethylene (trichloroethene) - HAP/VOC	2.8	A, B
Vinyl chloride - HAP/VOC	7.3	A, B
Xylenes - HAP/VOC	12	A, B

A – hazardous air pollutants

B – considered volatile organic compounds

On the other hand, in case of high methane content, LFG becomes an evidently valuable energy resource, as it is then able to sustain the fuelling of engines producing electricity and thermal energy.

Usually, the calorific value of landfill gas depending on CH<sub>4</sub>, and H<sub>2</sub> content ranges between 20000-25000 kJ/Nm<sup>3</sup>. It can be used as a supplementary or primary fuel to increase the production of electric power, as a pipeline quality gas and vehicle fuel, or even as a supply of heat and carbon dioxide for greenhouses and various industrial processes. Reported technologies that utilize LFG include i.a. internal combustion engines, gas turbines, fuel cells and boiler systems.

## 2.2. Landfill leachate

Leachate has four principal constituents; nutrients, especially nitrogen, volatile organic compounds, heavy metals, and toxic organic compounds. Nitrogen in the form of NH<sub>3</sub>-N has been identified as one of the priority components requiring removal to reduce leachate toxicity.

During XX century the “Dilute and disperse” landfill leachate treatment technology have been routinely commissioned at landfills within Europe. Landfill pits were generally unlined and generated leachate passes out into the wider environment where it mixes with ground waters and disperses. Dilute and disperse sites are no longer commissioned but have left a legacy of sites all over the Europe which could pose a contamination risk, particularly in sites located in areas with a shallow groundwater table. The EC Groundwater Directive was introduced in 1980 and classes compounds into List I (Black List) or List II (Grey List) depending on their polluting potential. List I and II compounds include NH<sub>3</sub>-N, NO<sub>3</sub> (nitrate), organohalogens, organophosphates, Hg (mercury), Cd (cadmium), Zn (zinc), Cu (copper) and Pb (lead), many of which are commonly found within leachate. Since 1993, under EC Water Quality Directives on groundwater protection and landfill, it has been legally required to have an impermeable geotextile lining for landfill pits. The need to assess the potentially harmful effect of wastes is expressed within the European Union in the Hazard Waste Directive 91/689 Code H14, part of the European Hazard Waste directive 94/904. This waste

directive is also in accordance with the landfill directive EU/31, 1999, which requires the assessment of waste classification and quality monitoring of leachates.

Research during the 1980's that examined water-courses receiving leachate found that attenuation was highly variable depending on the biological, physical and chemical processes within the landfill and the surrounding geological and hydrogeological environment (Williams 1999). Physical attenuation processes associated with dilute and attenuate landfill sites include adsorption, filtration and dilution. Chemical reactions take place between the leachate and surrounding environment. Ion-exchange occurs between cations and anions within the leachate and the soil and rock reducing contaminant mobility. Metals may precipitate forming hydroxides, carbonates, hydroxides and sulphides; acidic conditions solubilise metals while alkaline conditions induce precipitation. Oxidation and reduction reactions occur between inorganics. Some elements occur in more than one form. Iron for example interchanges between  $Fe^{2+}$  and the oxidised  $Fe^{3+}$ . The interchange between chemical forms can greatly affect the bioavailability of compounds. If leachate reaches groundwater in significant quantities a leachate plume following the direction of water flow develops. The extent of its dispersion, adsorption and absorption then depends on the surrounding geological environment. Leachate plume migration has been the focus for much research into landfill sites and leachate production. Fatta *et al.* (1999) described a Hydrological Evaluation of Landfill Performance (HELP) model, which has been developed by the US EPA to estimate the quantity of leachate produced from a landfill. The model, based on weather conditions and waste input, provides an indication of possible contamination from leachate plumes. At any landfill site, the extent of leachate production and plume migration is largely dependent on the hydrological balance of precipitation, surface runoff, evapotranspiration and infiltration.

The two factors characterizing a liquid effluent are the volumetric flow rate and the composition which in the case of leachate are related.

### **2.2.1. Leachate production**

Rainfall is the main contributor to generation of leachate. The precipitation percolates through the waste and gains dissolved and suspended components from the biodegrading waste through several physical and chemical reactions. Other contributors to leachate generation include groundwater inflow, surface water runoff and biological decomposition. Liquid fractions in the waste will also add to the leachate as well as moisture in the cover material. Moisture can be removed from the landfill by water consumed in the formation of landfill gas, water vapor removed in the landfill gas and leachate leaking through the liner.

So, the leachate flow rate is closely linked to precipitation, surface run-off, and infiltration or intrusion of groundwater percolating through the landfill. Landfilling technique (waterproof covers, liner requirements such as clay, geotextiles and/or plastics) remains primordial to control the quantity of water entering the tip and so, to reduce the threat pollution. The climate has also a great influence on leachate production because it affects the input of precipitation and losses through

evaporation. Finally, leachates production depends on the nature of the waste itself, namely its water content and its degree of compaction into the tip. The production is generally greater whenever the waste is less compacted, since compaction reduces the filtration rate.

### 2.2.2. Leachate composition

Landfill leachate can be characterized by very high concentrations of ammonia-nitrogen, almost always in excess of 1000 mg/dm<sup>3</sup>, often up to 2000 or 3000 mg/dm<sup>3</sup>, and sometimes higher. The chemical oxygen demand (COD) values are generally between 2000 and 8000 mg/dm<sup>3</sup>, although measured biological oxygen demand (BOD<sub>5</sub>) values are much lower – usually below 1000 mg/dm<sup>3</sup>.

There are many factors affecting the quality of leachates, i.e. age, precipitation, seasonal weather variation, waste type and composition. In particular, the composition of landfill leachates varies greatly depending on the age of the landfill. There are three types of leachates have been defined according to landfill age (Table 2). As landfill age increased, organics concentration (COD) in leachate decreased and increase of ammonia nitrogen concentration.

Table 2

Landfill leachate classification (Wang, Shen 2000)

Parameter	Young landfill	Medium landfill	Old
Landfill age [year]	<1	1-5	>5
pH	<6.5	6.5-7.5	>7.5
COD [gO <sub>2</sub> /dm <sup>-3</sup> ]	>15	3.0-15	<3.0
BOD <sub>5</sub> /COD	0.5-1	0.1-0.5	<0.1
TOC/COD	<0.3	0.3-0.5	>0.5
NH <sub>3</sub> -N [mg/dm <sup>-3</sup> ]	<400	400	>400
Heavy metals [mg/dm <sup>-3</sup> ]	>2.0	<2.0	<2.0
Organic compounds	80% VFA	5-30% VFA+HA+FA	HA+FA

In young landfills, containing large amounts of biodegradable organic matter, a rapid anaerobic fermentation takes place, resulting in volatile fatty acids (VFA) as the main fermentation products. Acid fermentation is enhanced by a high moisture content or water content in the solid waste. This early phase of a landfill's lifetime is called the acidogenic phase, and leads to the release of large quantities of free VFA, as much as 95% of the organic content. As a landfill matures, the methanogenic phase occurs. Methanogenic microorganisms develop in the waste, and the VFA are converted to biogas (CH<sub>4</sub>, CO<sub>2</sub>). The organic fraction in the leachate becomes dominated by refractory (non-biodegradable) compounds such as humic substances. Landfill leachates from old sites are usually highly

contaminated with ammonia resulting from the hydrolysis and fermentation of nitrogen containing fractions of biodegradable refuse substrates (Renou *et al.* 2008).

The existing relation between the age of the landfill and the organic matter composition may provide a useful criteria to choose a suited treatment process. In general, leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts.

The characteristics of the landfill leachate can usually be represented by the basic parameters COD, BOD, the ratio BOD/COD, pH, Suspended Solids (SS), VFA = Volatile Fat acids. HA = Humic Acid. FA = Fulvic Acids, Ammonium nitrogen ( $\text{NH}_3\text{-N}$ ), Total Kjeldahl Nitrogen (TKN) and heavy metals.

### **2.2.3. Detailed characterization of landfill leachate**

Landfill leachates contain a large number of trace compounds, some of which can be expected to create a threat to health and nature if released into the natural environment. A couple of 100 compounds have been identified in landfill leachates. Many of the reported compounds are not hazardous to health and nature as they represent degradation products ranging from small volatile acids to refractory fulvic and humic-like compounds. Some hazardous compounds have been detected, however, including aromatic compounds, halogenated compounds, phenols, pesticides, heavy metals, and ammonium.

Many of the compounds detected have been quantified at very low concentrations, often at nanogramme level. A large number of compounds can be expected to be present at concentrations below the quantification limits of standard analytical methods. Low concentrations do not eliminate environmental threats, as many compounds can be assumed to be hazardous even in small amounts and negative effects are often caused by multiple and synergistic effects. As the number of compounds identified in products is on the order of 100,000 and many of these, together with their transformation products, can be expected in landfill leachates, the amount identified is only a fraction of those present.

Usually, following types of compounds have been investigated: aliphatic compounds, mono- and polycyclic aromatic compounds, phenols, phthalic esters, ethoxylates, bromated flame retardants, polychlorinated biphenyls (PCBs), polychlorinated dibenzo-p-dioxins (chlorinated dioxins), polychlorinated dibenzofuranes (chlorinated furanes), polychlorinated alkanes (chloroparaffins), pesticides, organic tin, organic mercury, metals and other elements, sulfonamides and barbiturates, benzene and naphthalenesulfonates, methyl-tert-butylether (MTBE), dioxanes and dioxolans, phthalic acid monoester, ortho-phthalic esters and phthalic acid. The presence of following contaminants in landfill leachate was investigated: halogenated aliphatic compounds (dichloromethane, cis-1,2-dichloroethylene), benzene and alkylated benzenes (benzene, toluene, ethylbenzene, xylenes, n-propylbenzene, tert-butylbenzene trimethylbenzenes),

phenol and cresols, a bi-aromatic compound (naphthalene), phthalic esters, (diethyl phthalate, di-isobutyl phthalate, di-n-butyl phthalate, butylbenzyl phthalate, di-(2-ethylhexyl)phthalate), chlorinated benzenes (monochlorobenzene, dichlorobenzenes, trichlorobenzenes), chlorinated phenols (monochlorophenols, dichlorophenols, tetrachlorophenols) and pesticides (lindane, 2,4-D, MCPA, MCPP, 2,4,5-T, 2,4-DP, bentazone). The results of these research is shown in papers (Christensen *et al.* 2001, Kjelsen *et al.* 2002, Öman 2008).

It can be also noted that pathogens in leachate, toxicity, and estrogenicity also pose a threat to the environment.

Concentration of heavy metals in landfill leachate is usually at very low level. It is due to immobilization through sorption and precipitation. Most of the heavy metals deposited remain inside the landfills, and it has been suggested that <0.02% is leached out within the first 30 years (Flyhammar 1997). When mobilization occurs, it is through complexation to mobile inorganic and organic ligands and sorption to mobile inorganic and organic colloids.

#### **2.2.4. Landfill leachate toxicity**

Landfill leachate is a complex cocktail of constituents including dissolved organic matter, inorganic macrocomponents, heavy metals and a wide range of xenobiotic organic compounds of which over 400 have been identified. Large number of the compounds present in landfill leachates are hazardous and toxic to human health and the environment. Hazard assessment of landfill leachates is traditionally based on evaluation of individual chemicals through chemical analysis. This approach is important but has a number of limitations. Firstly, some chemical contaminants may occur below detection limits of chemical analysis and therefore isolation may prove difficult due to analytical technique limitations. A continuous chemical sampling regime may be required to detect changes caused by periodic discharges but this is costly and labour intensive. Finally, and possibly most importantly, chemical techniques are incapable of predicting pollutant effects on a receiving ecosystem. Through studying the response of biological organisms upon exposure to toxicants, ecotoxicology can provide an alternative or complementary method to that of traditional chemical analysis. In contrast to chemical analysis, a toxicity bioassay can integrate the biological effects of all the compounds present and also the bioavailability of substances. The occurrence of synergism and antagonism between chemicals is also directly reflected in bioassay results.

All chemicals have toxicity potential if they are present at high enough quantities, ecotoxicology can be used to identify the relationship between the quantity of the chemical to which an organism is exposed and the nature and degree of the harmful effects elicited. The relative toxicities of chemicals are generally related to their concentration, persistence and bioavailability in the environment.

Ecotoxicology has been employed successfully for the study of single chemical toxicity, but one of its most important applications is for the study of complex

effluents such as leachate. Within complex effluents several types of interactions between chemicals can occur. Chemical interactions can be described according to their resulting effect on overall toxicity. The additive effect occurs when the combined effect of chemicals is equal to the sum of the effects of all individual chemicals. This is generally assumed to be applicable to complex effluents, however this does not always work. If the combined effect of the chemicals is greater than the sum of the chemicals alone this is described as synergism or potentiation. Conversely, antagonism results in a less than expected toxicity and can result from a range of chemical and physical interactions between compounds. Four different antagonistic reactions are recognized; functional antagonism occurs where chemicals counterbalance each other; chemical antagonism is where chemical constituents react to produce a less toxic product; dispositional antagonism is where the chemicals' toxic effect is decreased through absorption, biotransformation or distribution; receptor antagonism occurs when chemicals bind to the same receptor binding site responsible for the toxic action thereby reducing overall toxicity. Chemical analysis can quantify leachate constituents but it cannot predict the interactions occurring between them, which may increase or decrease toxicity.

It is clear that chemical analysis alone provides limited information on the environmental fate of complex effluents such as leachate. Ecotoxicology can provide a bridge between traditional chemical analysis and complex field based studies through the use of biological organisms under controlled environmental conditions using defined principles.

Assessment of landfill leachate toxicity using bioassays has previously been undertaken for a variety of reasons, including the identification of toxic components within leachate, the impact of leachate on a receiving ecosystem, and the assessment of leachate remediation efficiency.

One of the earliest studies involving leachate toxicity was that of Cameron, Koch (1980), who highlighted the inadequacies of traditional chemical testing and, by utilising the recently developed *O. mykiss* assay, set a precedent for future investigations of leachate toxicity. This study identified the main components of leachate toxicity to fish as un-ionized ammonia, tannins and copper. It also highlighted the importance of testing leachate without pH alteration as this can affect the bioavailability of some of the leachate components. Later studies also established a link between ammonia and toxicity to a range of organisms including invertebrates and vascular plant species.

Ammonium can have a negative environmental impact through toxicity. Ammonia (NH<sub>3</sub>) standing in chemical equilibrium with ammonium (pK<sub>a</sub> 9.24 at 25°C) has proved to be very toxic, both acute and subacute, to aquatic organisms. The acute toxicity (LC<sub>50</sub>/EC<sub>50</sub>) to fresh water invertebrates has been found to vary between 0.53 and 22.8 mg NH<sub>3</sub>/dm<sup>3</sup> in tests of 18 invertebrate species, representing 14 families and 16 genera. The acute toxicity (96 h LC<sub>50</sub>) to fresh water fish varied between 0.083 and 4.60 mg NH<sub>3</sub>/dm<sup>3</sup> in tests of 29 fish species, representing nine families and 18 genera. The acute toxicity to saltwater invertebrates was found to

vary between 0.38 and 37 mg NH<sub>3</sub>/dm<sup>3</sup> (five species representing five families), and the acute toxicity (LC<sub>50</sub>/EC<sub>50</sub>) to salt water fish varied between 0.690 and 2.38 mg NH<sub>3</sub>/dm<sup>3</sup> (two species). Thus, landfill leachates have the potential of being acutely toxic to both fresh and salt water organisms. This has been confirmed in previous studies, which have reported ammonia to be the primary cause of toxicity of municipal landfill leachates (US EPA 1984, 1989).

In addition, ammonia is lost from leachate ponds through volatilization: the Henry's law constant is 32.2 Pa m<sup>3</sup>/mol at 20/25°C, and elevated ammonia concentrations in air may cause acute effects on vegetation.

The impact of the leachate on living organisms is serious and governed by several factors, such as heavy load of ammonia nitrogen (especially in un-ionized form), organic matter, heavy metals. Comparative toxicity assessment studies have revealed also the mutagenic properties of landfills leachate. Helma *et al.* (1996) showed that genotoxic potential of municipal solid waste landfill leachate was higher of that for effluents from pulp production, industrial wastewater, contaminated surface and groundwater or even drinking and bathing water samples. Leachate also can act as an endocrine disrupter and inhibit the embryos hatchability.

Given landfill leachate characteristic clearly shows the necessity of implementation of high quality landfill leachate management strategy (collection, retention, recirculation and final treatment prior discharging into the environment. This is the end-pipe strategy of managing pollutants.

### **3. Landfill fires – a source of pollution**

Landfill fires cause significant environmental impacts due to the emissions of toxic substances into the air, soil and water. The emitted odors and smokes are a nuisance for the neighboring communities and may threaten the health of those who are exposed to these conditions. The risk factor depends on the type of the burning wastes, the geographic location of the landfill site and the type of fire. Thus, the smoke emitted from a landfill fire may contain dangerous toxic gases, as CO, H<sub>2</sub>S, CH<sub>4</sub> etc., as well as carcinogenic substances as dioxins. Very high concentrations of dioxins in the goat meat and milk were confirmed for a specific farm 500 m from the landfill where fires took place. The problems of high dioxin levels, teratogeny, and ailing animals were confined especially in the goats of the farm and not in the sheep due to their grazing habits.

### **4. The influence of modern waste management strategies on emissions from landfills**

Modern strategies in waste management and landfilling techniques may decrease the impact of landfills on environment, by reversing the end-pipe strategy, and manage the pollutants: biogas and landfill leachate before they arise.

The European Council Directive on the Landfill of Wastes 1999/31/EEC (LFD), places a requirement on member states to draw up strategies to ensure

the amount of biodegradable municipal waste deposited at landfill progressively reduces over a 15 year period to only 35% of the total amount produced in 1995. The LFD also requires member states to only landfill wastes that have been subjected to treatment and incineration, leading to a reduction in their quantity or hazard to human health or the environment. It is anticipated that Mechanical and Biological Pretreatment (MBP), and incineration of municipal waste will increase substantially in the Europe, and Poland as well, in order to achieve these objectives.

#### **4.1. Mechanical-biological pre-treatment of waste (MBP)**

Why is a reduction in the landfilling of biodegradable organics implemented in the EU Landfill Directive and what are the main advantages of applying mechanical-biological pre-treatment (MBP)?

By undertaking MBP, processes which normally take long periods of time (decades) to complete in a landfill are shortened to several months. The emissions potential of the waste is reduced to a great extent during pre-treatment so that, compared to non-pretreated waste, a significant reduction in emissions occurs. The production of “compost” to be applied in agriculture or horticulture is not the aim of mechanical-biological pre-treatment, because the content of heavy metals and other harmful or interfering substances is generally too high.

Mechanical and biological pre-treatment of MSW has been required through national legislation in a number of EU member states for several years. The perceived benefits of such pre-treatment are:

- Reduction in the strength of leachate produced, and the quantity of landfill gas generated;
- Reduced clogging of leachate drainage systems;
- Improved waste settlement characteristics, and a shorter timescale to waste stabilisation.

MBP MSW reduces the emissions regarding organics and nitrogen as well as the gas production by approximately 80-90%. Due to MBP of the MSW, the landfill characteristics achieved with this waste (MBP material) would also be significantly different from raw MSW in the following ways:

- The separation of the high calorific value fraction (e.g. plastics, paper, wood) means MBP material has a low structural material content.
- Active gas extraction would not be necessary since the gas formation potential is low; however, the residual amount of gas produced would need to be biologically oxidised before leaving the landfill.
- By adjusting the waste properties (e.g. water content) and using appropriate landfilling techniques, the MBP material would have a high density ( $\rho \pm 1.5 \text{ t/m}^3$  wet weight) which may result in low permeability ( $k_f \pm 10^{-8} \text{ m/s}$ ) and savings in landfill volume.
- The mechanical stability of the landfill would be affected by the water content of the waste material and the way the landfill is operated. If for

instance the pore gas pressure or the pore water pressure is too high, landfill stability may be reduced.

Due to the complex procedures of MBP waste landfilling during rainfall should be avoided or other options may be advantageous. A proposal for the construction and operation of bale landfills has been made already back in 1993. This type of landfill is especially suitable for mechanically-biologically treated wastes as the relatively homogenous MBP material is compacted to high density bales and subsequently installed (stacked) at the MBP landfill. An example of that landfilling technique is given on Figure 2.



Fig. 2. An example of landfilling of balled waste on landfill in Karcze, near Sokółka, Poland (phot. A. Białowiec 2010)

Landfilling of MBP municipal solid waste has also possible impact on the environment, and the strength of it should be investigated.

Some research projects to consider the potential impact of the Council Directive, on the quality of leachate that will be produced in future from landfills, have been started.

There are two broad categories of organic residues from waste pre-treatment:

- mechanically sorted organic residues,
- and biologically stabilized MSW or MSW fractions.

Mechanically Sorted Organic Residues (MSOR) (also termed “residual wastes”) are fine fraction residues from a mechanical sorting process, which cannot

be reused or recycled. The maximum size fraction of MSOR typically passes through a 40 mm or 100 mm screen, and the quality and proportion of MSOR will depend on the extent to which:

- Wastes have been subject to source separation, e.g. of garden and kitchen wastes;
- Wastes have been separated for recycling at a Materials Recovery Facility.

Typically, the larger-size fractions discarded during the sieving process are of higher calorific value, and are diverted for either direct incineration (preferably with energy recovery), or for the production of refuse-derived fuels.

The high organic content, high moisture content, and small particle size of the MSOR appears to give rise to much higher landfill gas production rates and stronger leachates. If this material is landfilled, particularly in isolation from other waste streams, then extremely strong leachates must be expected, that will persist for as long as, or longer than, those from conventional MSW landfills. With this very high pollution potential in mind, MSOR wastes have often been subjected to various composting processes, for many years in some European countries.

Many detailed studies have demonstrated the very strong leachates that are generated when untreated MSOR are landfilled. In leachate arising from a landfill containing of MSOR, the contaminant concentrations values may reach levels as follow: COD=60,000 mg/dm<sup>3</sup>, BOD<sub>5</sub>=40,000 mg/dm<sup>3</sup>, Kjeldahl-N=4,000 mg/dm<sup>3</sup>, and Cl=6,000 mg/dm<sup>3</sup>.

Other studies have looked at the benefits of composting MSOR, in terms of reductions in pollutant emissions. Leikam, Stegmann (1999) studied the behaviour of composted MSOR wastes in landfill simulation tests, in comparison to untreated MSOR. For treated MSOR, the acetogenic phase during which strong organic leachate is produced was absent, and after about 250 test days the COD of the leachate was below 1,000 mg/dm<sup>3</sup> (BOD<sub>5</sub> <20 mg/dm<sup>3</sup>). A much more significant benefit of pre-treatment becomes apparent when concentrations of total-N (primarily ammoniacal-N) are considered. Whereas the total-N content in leachate from untreated residual waste stabilised at about 1,000 mg/dm<sup>3</sup>, this value was below 200 mg/dm<sup>3</sup> for pre-treated wastes.

Key findings from these and other published studies on leachates from landfilled MBP wastes are that:

- Organic residues from mechanical sorting (MSORs) can produce leachates with higher pollution potential than both acetogenic and methanogenic leachates from conventional landfills;
- Composting such residues can reduce the organic pollution potential from both leachate and landfill gas, through the avoidance of the peak acetogenic phase of decomposition;
- Concentrations of ammoniacal-N in MBP leachates can be either similar to, or much lower than, methanogenic leachates from conventional landfills. This raises the possibility that a nitrogen removal or attenuating process may operate, to varying extents, during composting;

- Landfills receiving MBP wastes will pose a risk to groundwater similar to conventional MSW landfills that have become methanogenic, and are therefore likely to require a similar period of time before active management and treatment of leachates ceases to be necessary.

## 4.2. Types of landfill

Several landfilling technologies will be presented, as an example of specific landfill evolution from the worst technology to the most friendly to the environment. Worldwide, open dump landfills have been recognized as unable to meet the sustainability target and are being replaced by more or less engineered landfilling systems. Technological measures have been introduced to achieve a better control over liquid and gaseous emissions from landfills. Control of leachate aims at preventing groundwater pollution whereas control of gas is meant to reduce emissions of greenhouse gases, prevent fire hazards, odours and vegetation damages.

The conventional landfilling scheme, typically based on anaerobic degradation of waste, makes use of bottom liner, top soil cover, gas and leachate collection and treatment systems and, lately, also gas utilization for energy recovery. Although these technical measures can significantly reduce the uncontrolled release of gas and leachate, the potential environmental impacts still remain high and able to threaten the environment far beyond the time frame of a generation. New “active” waste landfilling technologies have therefore been developed in the last couple of decades, including bioreactor and semi-aerobic technologies.

In bioreactor landfills the waste degradation process is microbially optimized to achieve a faster and deeper stabilization of waste. This is mainly accomplished through the recirculation of the collected leachate. Leachate recirculation ensures continuous supply of nutrients and moisture close to field capacity, two key conditions for promoting the degradation reactions together with leachate in order to flush-out the soluble waste components, in a process called “waste irrigation” or “waste flushing”. These landfills are referred to as flushing-bioreactor landfills. Besides waste irrigation, measures to reduce ammonia ( $\text{NH}_3$ ) concentrations in leachate are commonly taken in flushing landfills, such as leachate nitrification and in-site denitrification.

The semi-aerobic landfilling technology, developed in Japan, combines anaerobic and aerobic metabolism to stabilize the waste mass. The degradation mechanism may initially be anaerobically driven and enhanced by the leachate recirculation operation. This first step is kept active for 5 to 10 years, until the methane ( $\text{CH}_4$ ) generation of the relatively shallow landfill gets too low to justify gas utilization for energy recovery. The subsequent aerobic step is initiated by injecting air from the bottom of the landfill. A convective air flow will then proceed autonomously. Compared to anaerobic degradation, aerobic metabolism leads to faster waste stabilization rate, greater generation of landfill gas and lower leachate production.

### 4.3. Sustainable landfilling

Europe aims at moving up in the waste hierarchy, away from landfill and more and more towards a recycling and recovery society. Since 1999 the Landfill Directive contains requirements to reduce the amount of waste to be landfilled. In 2016 member states are not allowed to landfill more than 35% of the biodegradable municipal waste they landfilled in 1995. Some countries are allowed to reach this target four years later (see Poland). The key objective of the waste policy is to prevent the generation of waste. If prevention is not possible, waste should be re-used or recycled. If that is not possible either, it should be incinerated with energy production. Only if other waste treatment options are not suited, the waste should be landfilled.

No matter how much prevention, re-use and recycling a society manages to realise, there will always be a role for landfill in a waste management system. It will not be economically sound to have enough capacity to recycle or recover all waste under all conditions. While aiming for more prevention it is economically unsound to invest in recycling and recovery of waste that is bound to disappear. Furthermore, the amounts of waste fluctuate over the year. Sometimes the amount of waste exceeds the capacity for recycling, recovery or incineration. Not all wastes can be recycled, recovered or incinerated. For some wastes landfill is the best option. And in case a recovery or incineration plant is out of operation because of maintenance, repair or an accident, the waste should not remain in the streets. This means that even in a recycling and recovery society some wastes need to be landfilled. The landfill sites are the ‘safety net’ in a good waste management system. Landfill should be carried out in a way such that future generations do not have to worry about it. It should be done in a sustainable way.

Isolation of landfills by means of impermeable liners seems to become the standard in Europe. Isolation stops all processes in the landfill. Liners may hold for fifty years. They may even hold for five hundred years. But they will inevitably fail at some point in time. When the liners fail the processes, as driving force for emission, will start up again. Therefore potential emissions are postponed to future generations. Aftercare is a requirement of many national regulations. Aftercare generally has to be carried out for at least thirty to sixty years after the closure of a landfill. Some countries require it ‘as long as the competent authority considers it necessary’. Clearly aftercare is always required for longer than one generation. An intrinsically safe solution would be more sustainable. A society striving for sustainable development consequently needs sustainable landfills.

There is no internationally accepted definition of sustainable landfill. With respect to landfills very often terms as stability, completion, end-point and threat to the environment are used together in discussions about sustainability. According to Scharff *et al.* (2007), a selection of definitions is:

- SWANA Stability Subcommittee: A landfill is ‘functionally stable’ when the waste mass, post-closure, does not pose a threat to human health and the environment. This condition must be assessed in consideration of leachate quality and quantity; gas composition and production; cover,

side-slope and liner design; site geology and hydrogeology; climate; potential receiving bodies, ecosystems and human exposure; and other factors deemed relevant on a site-specific basis.

- Anglo-Welsh Environment Agency: Completion is defined as that point at which a landfill has stabilised physically, chemically and biologically to such a degree that the undisturbed contents of the site are unlikely to pose a pollution risk in the landfill’s environmental setting. At completion, active aftercare pollution controls (e.g. leachate management and gas management) and monitoring systems are no longer required.
- DHI: Waste at final storage quality provides a situation where active environmental protection measures at the landfill are no longer necessary and the leachate is acceptable in the surrounding environment.
- Technical University of Hamburg: The aftercare phase may end when the emission potential is that low that the actual emissions do not harm the environment.

Although the different definitions use slightly different wording, there seems to be a general consent that a sustainable landfill or a landfill for which it is considered safe to end aftercare is a landfill that within a limited period of time reaches a state where the undisturbed contents no longer pose a threat to human health and the environment. At that point, often called completion, aftercare can be ended. It is important to note that this is in accordance with the intention of European waste legislation. The Annex II of the Landfill Directive requires neither isolation nor aftercare for landfills for inert waste. And it defines inert waste in similar terms. That is: not posing any threat to human health and the environment.

Throughout the world various methods are proposed to stimulate landfill processes in order to accelerate achievement of a stabilised landfill. In the end the goal is the same: to achieve completion of the landfill. Most of the proposed methods relate to pre-treatment or bioreactor type of operation for biodegradable organic waste. When most of the organic carbon has been degraded the behaviour of the landfill body converges to the behaviour of an inorganic waste landfill. The same applies to (suitable types of) hazardous waste after immobilisation or cement stabilisation. In other words: for all types of waste on the road to completion it seems inevitable to ‘pass through’ a stage that is comparable to an inorganic waste landfill. An inorganic waste landfill is not necessarily a stabilised landfill. There are components that cannot be biologically stabilised or immobilised with cement-like additives. Such components either need to be flushed out or ‘caught’ in ‘precipitates’. When that state is achieved the behaviour of the landfill is very much like the behaviour of a landfill for inert waste.

In landfills, containing organic material, biodegradation is the most important process. It produces, removes, mobilises and immobilises key pollutants. Biodegradation can be enhanced by leachate recirculation and leads to reduced emissions. The flushing of contaminants from the landfill appears to be the limiting factor. Emissions of contaminants from inorganic waste can meet the criteria for inert waste, except for chloride and sulphate. Clever combinations of waste enable

us to create beneficial conditions for the leaching behaviour of contaminants. Oxyanions and salts seem to be critical parameters in the judgment of stabilised waste with respect to the criteria for hazardous waste. Although not all questions have been solved in detail, it can be stated that most processes are known and applicable. It is estimated that applying the processes would result in a cost increase of 10 to 20% compared to the costs of an EU Landfill Directive compliant landfill.

The sustainable landfill:

- has final emissions that are lower than ordinary landfills;
- has an emission release that occurs within a shorter period of time;
- enables active control and prediction of emissions and;
- is technically and economically feasible.

## **5. Benefits from waste landfilling**

Earlier, most common threats arising from waste landfills were described. In this place it is time to re-balance the arguments in favour of the benefits of restricted waste disposal to landfill. In particular, when considered in a long term, landfilling may convert a difficult part of the waste stream to short-term provision of gas, may provide long-term carbon storage, and landfill afteruse may promote brownfields redevelopment resulting in new opportunities for landscape and environmental improvement.

Waste management options are not interchangeable. The various strategies available for solid waste management are generally considered as a hierarchy of sustainability opportunities with waste reduction at source as the best option and landfilling as the worst. However, at present, alternative waste disposals are subject to criticism either by environmentalists or by landfill operators and contractors.

The ideal ‘zero waste’ option is often discussed, but concrete initiatives are still unfeasible even for the most industrialised countries. Recycling is difficult to be put into practice efficiently and at a reasonable cost. In particular, it requires efficient separation of different waste fractions both at the sources and after collection. Waste segregation of organic fraction, either biodegradable or combustible, has been identified as significant sources of various pathogens. Composting, the most valuable way to recycle organic waste, has also health implications for both workforce and residents in proximity of large scale composting plants. Incineration provides the main alternative to landfill but presents another range of associated problems. Its technology is very expensive, in order to deal with all the potential hazards, in particular, air emissions, it is subject to criticism for possible hazardous emissions, failure to eliminate pathogenic agents or failure to immobilise heavy metals or else poor waste management economics; furthermore, public perception of risk appears to greatly exceed actual risk. Bottom ash residues released from municipal waste disposal incinerators may be disposed of to landfills, with or without pre-treatment, but inhalation risks from this source in landfills have been assessed as being insignificant (Luria, Dickinson 2007).

Landfilling has the lowest internal costs. Its externalities (health, social and environmental detriments computed on an economic base) are traditionally indicated as significant. However, their quantification is questionable and, again, any consideration does not take into account new available technologies.

## **6. Evidence base linking landfills with environmental health**

Whilst landfill remains the dominant waste management option for municipal wastes, there is a shortage of landfill voids to accommodate new landfills in many countries. Modern sanitary landfill design has sought an engineering solution for safe containment of waste that has moved toward dry entombment of waste, on the assumption that anaerobic degradation processes are sufficient for the waste eventually to stabilise to an inert state. There is an argument that this strategy is flawed largely because the lack of water ingress slows the rate of waste dilution and degradation, leaving potential problems for the future whilst uncertainties also remain about the long-term integrity of HDPE and other liners. Furthermore, increased confidence and reliance on lining systems has meant that geological and hydrological conditions of the landfill location may be given less precedence at the planning stage. It has been suggested that landfill sustainability would be improved by mechanical and biological pre-treatment involving mining, aeration and flushing.

Landfilling technologies suffer from a general stigma in the eyes of both the public and public authorities, mainly related to safety and public health. The negative viewpoint of the safety of landfills is probably distorted, and any link between landfilling and public health is controversial but unproven.

In general, the fear of risk of water (regardless groundwater or not) and air contamination dictates the others. Most of the public thinks that there is no such thing as a secure landfill and that all landfills eventually leak. This is probably due to the fact that emission to water is a typical landfill risk. From a scientific point of view, any link between landfilling and health is quite controversial. There is some evidence that living near a hazardous waste site may contribute to the development of coronary heart disease and occurrence of acute myocardial infarction. However, detailed analysis of population-based health data has shown very few cases where environmental pollutant exposures, including those potentially associated with landfill sites, can be linked to adverse birth outcomes, including congenital birth anomalies, stillbirths or neonatal deaths (Luria, Dickinson 2007). An extensive study of people living within 2 km of 9565 UK landfill sites, operational from 1982 to 1997, showed no excess risk of cancers of the bladder and brain, hepatobiliary cancer or leukaemia (Jarup et al. 2002). The US EPA estimated virtually zero risk of cancer from 60% of 6000 MSW landfills, in 1993 (Chilton 1993). Thus, a causal link between human health is living close to landfill sites has not been demonstrated, despite some evidence of a correlation; there is a likely interaction of negative findings with other socioeconomic and environmental variables (Luria, Dickinson 2007).

## 7. Summary

Landfills emit pollutants to the environment. When they are wrongly designed, operated and closed, either the air receives the emission of greenhouse gases, trace components, and odors, or ground water, lakes, rivers, wetlands are polluted by strongly contaminated and toxic leachates. On the other hand, a landfill may have a range of benefits. It gives the opportunity to dispose some form of repository for non-recyclable and untreatable wastes; at contained costs with short-term provision of gas, and long-term carbon storage.

If associated to a well planned after-use, landfilling may lead to a ‘win-win’ approach to waste management, by improving local environment and landscape, contributing to economic and health regeneration of the area, and being based upon some simple actions.

In the programming and planning phase, we should highlight several factors including the need for some form of repository for non-recyclable and untreatable wastes, availability of the new landfill technologies, the costs of these technologies, and possibilities of low-value degraded landscapes using.

In the operating phase, we should make clear that casual – rather than causal link between landfill and health is evident, and also that the landfill is not the end-use of the site but a transition to a better use.

In the aftercare phase, we should promote shortening to medium term biogas provision ability, the capability of long-term carbon storage from waste. We should consider the better final use for the site linked with any social and health improvements in the local context.

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