Gas Emission from Landfills

An overview of issues and research needs

Survey

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1. Introduction and Summary

This report presents a brief overview of issues related to the management of solid wastes at landfills and the resulting impact on emissions to the air. It does not seek to give an extensive review of the field, but only to outline important issues and to indicate relevant research areas.

The discussion has been divided along three themes:

- the landfills and waste types,

- the (known) emissions,

- gas formation and emissions.

For each of the above topics conclusions and recommendations are presented at the end of the individual chapters.

The last chapter presents a synthesis of the previous chapters and also includes recommendations about what measures should be taken. In summary, the authors claim that:
- Gaseous emissions from waste management (including landfilling) are environmentally significant on a global scale today and will acquire more importance in the future.

- Gaseous emissions are also important for the working environment and for people living near landfills and other waste treatment facilities.

- The main problems associated with gas emissions from wastes are:

- human health hazards,

- contribution to greenhouse effect,

- odour problems,

- explosion and fire hazards.

- Recent trends in changes of material flows to landfills increase the complexity of the issue and hence the need for research.

- The areas where further R & D is needed include:

- characterisation of materials and their potential emissions,

- processes generating emissions (degradation and transformation processes),

- emission control (both active gas abstraction and passive treatments),

- emission prediction and evaluation tools,

- impacts of emissions on the working environment.

- In addition to research funding, there are also other measures which could contribute in a positive way to the development of knowledge and the technical level of operations within the field, such as:

- supporting networking through meetings and travel grants,

- refining and making more accessible the statistics on landfilling and waste treatment in general,

- stimulating the participation of waste management companies in R & D efforts.

2. Landfill and waste types

There are about 300 active landfills receiving more than 50 tons of waste per year in Sweden (excluding some landfills owned by the industry). In 1994, a total of 6 Mt were landfilled (Table 2.1). The amounts of wastes landfilled have been declining somewhat over the last decade and seem to have stabilised now. A further decrease may be expected after the introduction of the landfill tax in the year 2000, but an increase in conjunction with increased construction activity cannot be excluded either.

Table 2.1   Total amount (kt yr^{-1}) of landfilled waste in 1994 (Modified after RVF 1996 and http:www.rvf.se)

Waste category	1994	1995	1996	1997
Household and similar	1380	1200	1110	1150
Yard and garden	80	60	70	50
Construction and demolition	900	950	885	700
Energy extraction	660	680	700	675
Municipal sewage treatment	610	540	470	455
Industrial sewage treatment	190	190	205	210
Industrial, general	1060	1120	1050	970
Industrial, sector specific	490	470	435	405
Mineral recycling industry		10	1	2
Special	90	120	130	133
Other	620			

The main generic waste categories entering municipal landfills which may be expected to cause gas emissions are household wastes, ashes, construction wastes and sludges. Of these wastes, the household (and similar commercial) wastes are expected to generate the highest amounts of landfill gas, strongly dominated by the main components, methane and carbon dioxide. The other gas-generating waste categories could possibly release a higher proportion of various trace components. It should also not be forgotten that ashes and other alkaline wastes will absorb atmospheric carbon dioxide, and thus serve as a sink. Similarly the

methane emissions result in the establishment of active methane oxidising microorganisms in landfill covers, which may then also serve as a sink for atmospheric methane (Bogner *et al.*, 1997b).

An increasing activity at landfills is the intermediate storage and processing of recyclable materials and composting of yard and garden wastes. About a million tonnes of recyclable materials were placed in intermediate storage at municipal landfills in 1997 and more than 230 kt of wastes were composted at 118 landfills in 1997 (http://www.rvf.se). Emissions to air from the intermediate storage, sorting and composting activities are likely but little documented. The main emissions that could be anticipated to emanate from these practices are carbon dioxide, methane, hydrogen sulphide and nitrous compounds, but some wastes may also emit more hazardous organic compounds.

In recent years a "producer responsibility" principle has been enacted for packages and some other materials. At the moment whether to also include electronic wastes in this category is still under consideration. The management of these materials are organised by so-called "material companies" who purchase services from various entrepreneurs for the actual collection and handling of the materials, some of which is done at landfills. It is likely that the management of these recyclables will generate new opportunities for emissions to the atmosphere, but there is no data available to support or denounce this assumption.

A number of sector specific industrial wastes may cause gas emissions. One known major source of emissions are different organic wastes, *e.g.* from the paper industry. These waste are often in the form of sludges or relatively finely graded materials which are not easily degassed using conventional LFG systems. There are also degradable organic compounds in some other industrial wastes such as ashes and various filter wastes from air cleaning processes. Some industrial wastes also give off gas by chemical reactions upon wetting, for example lime and coke dust, carbide wastes, silica iron slag, some filter dusts, and spent potlining from aluminium production (Bergman, 1996). A large and increasing part of the material being landfilled is waste from various recycling operations. Some of it is deposited in municipal landfills and some in special landfills, for example car-scrapping wastes. The potential for atmospheric emissions from these wastes varies with the materials processed and should be assessed individually for each site. No comprehensive data is available today.

Landfill gas (LFG) was abstracted at 59 of the active landfills in 1997. At 34 of these landfills special cells were used for biodegradable materials, in order to improve the collection efficiency. A total of about 437 GWh of energy from LFG was used from the 59 active and some closed landfills equipped with gas abstraction systems. In addition LFG corresponding to about 9 GWh of heat was flared (http:www.rvf.se). Using a conversion factor of 10 kWh m^{-3} of methane gas, the total amount of energy released would correspond to an abstraction of about 45 Mm^3 yr^{-1} of methane and a similar amount of carbon dioxide. In effect, a greater amount is probably collected since all use and flaring might not be accounted for, *e.g.* internal use. The gas is in any case mainly abstracted from landfilled household waste, from which it

has been estimated that about 100-150 Mm³ of methane is formed per year (SNV, 1993, 1994). This would imply an abstraction efficiency of 30-50% of the potential, which seems to be a bit on the optimistic side, in view of the low average specific yield of the existing plants. In 1995, the average yield of Nordic LFG plants was less than 4 m³ of LFG per tonne of waste in place and year, which is about 20-25% of what was abstracted at Swedish test cells (Lagerkvist, 1997; Lagerkvist, 1997). All of the numbers cited are of course approximations. It is in particular difficult to assess the quality of the emission potential estimates published by the Swedish Environmental Protection Agency (SNV), since no method description and base data are given.

The landfills vary much in size and in technical level. The 25 largest municipal landfills receive about half the total amount of waste whereas the 175 smallest only receive about 10 percent (http:www.rvf.se). Since all of the major landfills already have gas collection systems installed, the average size of additional landfills where gas is abstracted tends to decline with each passing year, as illustrated by Figure 2.1. One could imagine that it would be more difficult to design and operate smaller landfills for minimal emissions. Based on the assumed dependency between the landfill size and gas collection efficiency, one could expect a still lower gas collection efficiency in LFG plants established in recent years. However the correlation between size and efficiency is very weak, as illustrated in Figure 2.2.
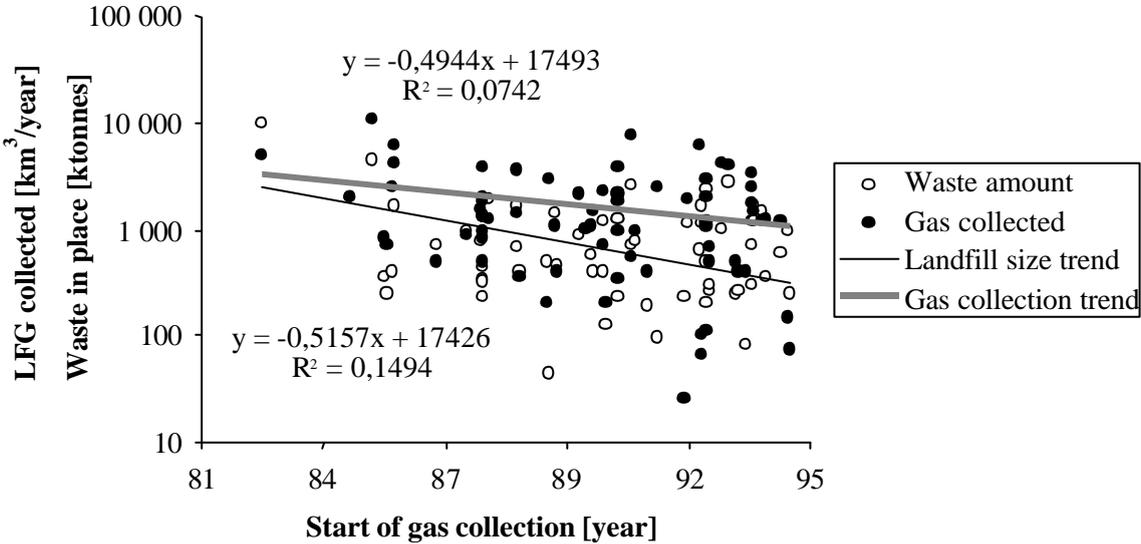
Figure 2.1   LFG plants established before 1995 where gas was used in 1994. Amount of waste being degassed and volume of LFG collected (from Lagerkvist 1997).

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The chart shows: y-axis "LFG collected [km³/year]" with values 0, 2000, 4000, 6000, 8000, 10000, 12000. x-axis "Waste in place [ktonnes]" with values 0, 2 000, 4 000, 6 000, 8 000, 10 000, 12 000. The trend line equation is:

$$y = 0{,}7571x + 1036{,}4$$
$$R^2 = 0{,}3085$$
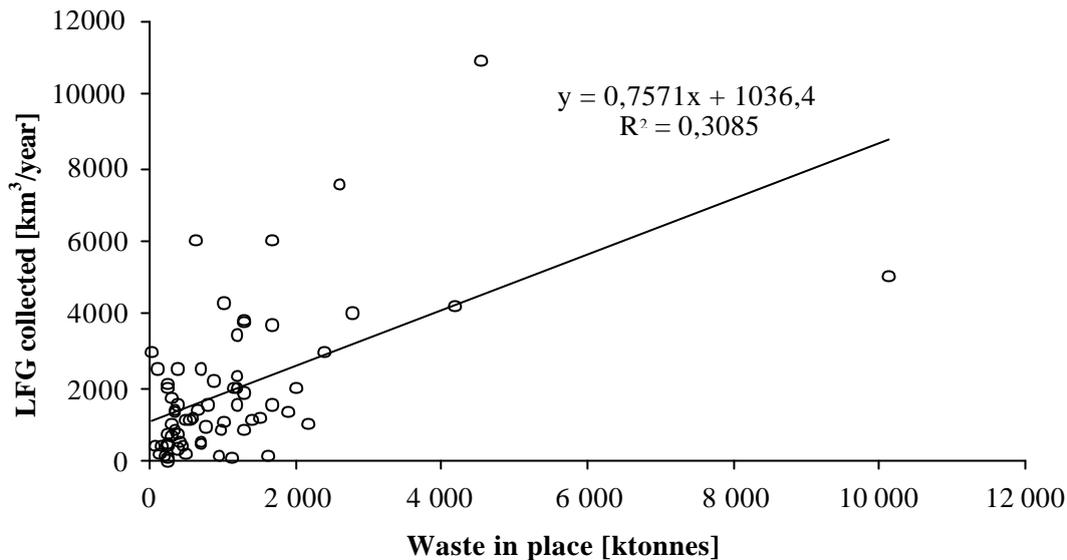
Figure 2.2   LFG plants established before 1995 where gas was used in 1994. Volume of LFG collected as a function of the amount of waste being degassed (from Lagerkvist 1997).

This indicates that fairly small plants can have a high efficiency and thus that the scope for development and improvement of the gas collection technology is substantial.

In view of the present trends and regulations (for example the new European Landfill Directive) one can predict a further decrease in the landfilling of untreated household waste and similar biodegradable wastes which will eventually result in a less gas being generated from such wastes. One must however remember the present potential, and substantial gas emissions are to be expected from existing landfills also 20 years into the future. Since investment decisions are based on expectations, there is a risk that some LFG collection systems that could have a fairly good collection efficiency will never be built. There is also a risk that the technical advances made over the last decades with regard to biocells and gas collection technology may remain under-exploited.

One can also predict that an increased landfilling of mechanical-biological pre-treated wastes and recycling wastes will take place. The amounts of materials which are handled in treatment and recycling operations and kept in intermediate storage for these purposes is also likely to increase, as are the possible emissions from these materials. The impact of this cannot be evaluated on the basis of the data available today. There are good grounds to improve the information about these wastes, since on the one hand a number of potentially very hazardous compounds may be emitted from some wastes, and on the other there is a great exposure and human health risk, in addition to the potential environmental impact.

Another class of landfills which may come into existence in the near future is the *in situ* stabilised ones. In order to stabilise landfilled waste, and contaminated soils, in situ aeration is employed today and this practice has been shown to have a reduce the potential of both gas and leachate emissions. In comparison to landfill mining combined with mechanical-biological or thermal treatment, the method is cost effective (Heyer & Stegmann, 1999) and may be expected to find increased application over the next decades. The treatment has in itself of course an emission potential, which will differ both qualitatively and quantitatively from that of an anaerobic landfill. There will also be some residual emissions generated after treatment. Since the method has been only sparingly documented at the moment, one cannot rule out the possibility that hazardous compounds may be generated/released, when specific types of wastes/contaminants are treated in this manner, so there is a need for further evaluation and testing of protocols.

Other major waste fractions, such as ashes and sludges, will probably remain much the same, while construction and demolition wastes will fluctuate with the economic situation as before. For all these wastes there is a lack of basic process data from landfills, and of a basis for emission estimates. For industrial waste landfills there is a need for evaluations on a case by case basis.

2.1 Conclusions

Landfills are an important element of waste management, both today and in the future. The complexity of landfills is increasing and there are many aspects which may affect emissions to air which are poorly documented today. A number of "new" waste types and waste processing or storing practices need to be characterised. much could be gained through a better implementation of the existing knowledge on gas generation and abstraction. There is also a need to develop better practices with regard to operation and emission control, in particular to better adapt the techniques to different waste materials.

3. Compounds emitted by landfills into the air

These emissions constitute one of the major environmental concerns regarding landfills. Gaseous compounds are produced following biochemical reactions, such as the methane and carbon dioxide generated in MSW landfills from the anaerobic degradation of the organic fraction of the waste, or they may be released by direct volatilization from sources already present in the waste. Particulates, in particular dust, and aerosols are other significant atmospheric emissions from landfills.

The gaseous compounds emitted from landfills have various impacts on their surroundings and act on different scales, as illustrated by Figure 3.1. In addition to having impacts over a large spatial scale, gaseous emissions also act on different time scales. Compared to most other processes used in waste treatment, those occurring inside the landfill and the emissions they generate extend over a very long period of time after the waste has been disposed: from tens to hundreds of years. Not only is the period of significant emissions long, but the compounds emitted will themselves have effects and life-spans of varying duration. Odours and dust, for example, are mainly transient phenomena, while some of the anthropogenic trace compounds in LFG may persist and accumulate in organisms or natural ecosystems over very long periods of time. Methane constitutes both a very short term and acute explosion hazard and has a much more far-reaching and long-term effect on global warming. In the following paragraphs, we will review some of these compounds, their origins, characteristics, impacts and relevance.
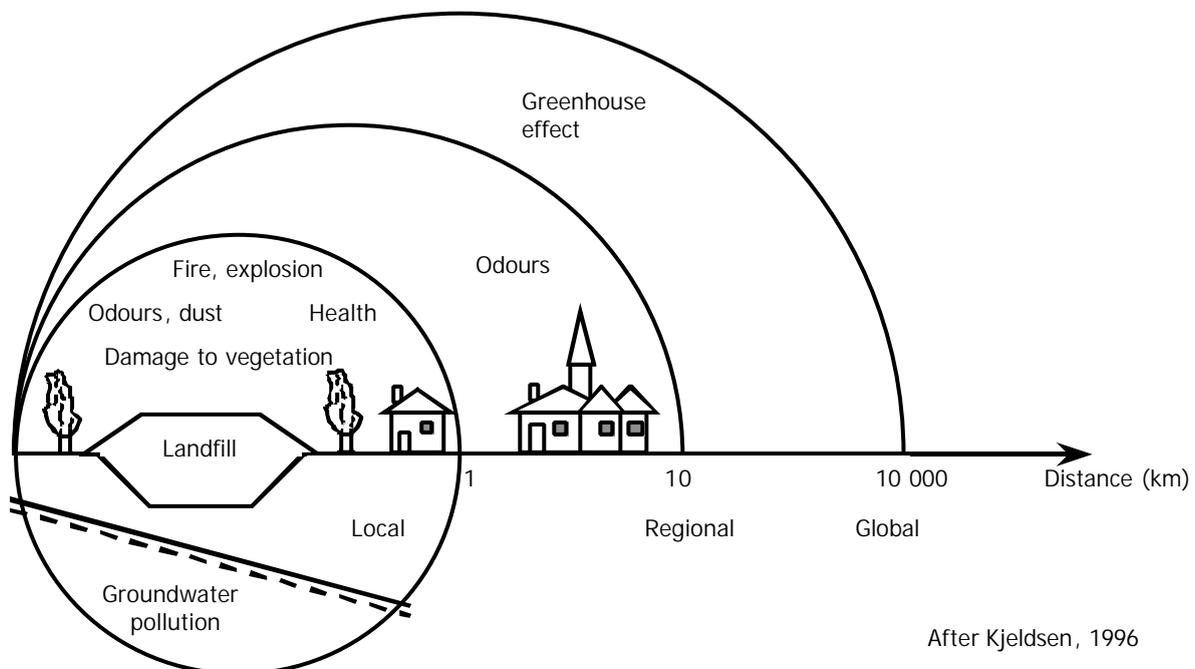

Figure 3.1   The different scales of the impacts of gas from landfills (after Kjeldsen, 1996).

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3.1 Carbon dioxide (CO_2)

Carbon dioxide is the main gaseous form of carbon. It arises as one of the waste products of the biodegradation of organic compounds, both aerobically and anaerobically. Alongside methane, carbon dioxide is one of the two principle components of the gas generated in MSW landfills, where it typically makes up between 20 and 50% of the gas.

Waste from energy abstraction from MSW (MSW incinerator residues) contain between 2 and 4 weight-% of organic carbon for bottom ash and 1 wt% for air pollution control (APC) residues. Microbial processes may therefore occur and degrade the organic compounds either aerobically or anaerobically, with a resulting production of CO_2. Heyer and Stegmann (1997) measured a small release of CO_2 in simulated deposits of bottom ash, while Belevi and coworkers (1993) observed microbial degradation of the organic carbon of incinerator residues during incubation experiments. CO_2 may also evolve from various kinds of industrial wastes.

Atmospheric CO_2 is a limiting factor in photosynthesis and is essential to plants. However, when present at high concentrations in soils, it can result in asphyxia due to oxygen displacement or even be directly toxic to plants. The normal CO_2 concentration in soils varies between 0.04 and 2%. An elevated concentration of CO_2 in a soil, a situation typical of landfill cover soils, is directly toxic to the plant roots, even if there is enough oxygen available. An indirect effect of carbon dioxide could be a lowering of the soil pH and the consequent changes in soil composition. Normal development can occur till 5%, though tolerance may vary between plant species (Maurice, 1998).

CO_2 is classified as an intermediary between toxic and non toxic substances. It acts by displacing oxygen in the respiratory system. Its ambient concentration lies around 250 - 350 ppm. The Threshold Limit Value (Nivågränsvärde - NGV) for CO_2 is 0.5%, with a Short Term Exposure Limit of 1%. At 3%, breathing becomes laboured and headache or drowsiness develop. Above 5%, it represents an acute danger for life.

Carbon dioxide must therefore be considered a serious safety threat in landfill environments. Several deaths due to CO_2 asphyxia have occurred on or near landfills in recent years, in underground or closed environments such as drains or culverts, where landfill gas had accumulated. In LFG treatises this hazard is generally treated in far less detail than the explosion and fire hazards due to methane (see *e.g.* Christensen, *et al.* eds, 1996; Gendebien *et al.*, 1992). It is interesting to note however, that a ten times higher dilution is required to lower the CO_2 concentration in pure biogas to below the NGV (0.5%), than is required so as not to exceed the lower explosivity limit (LEL) for methane (5%). Requirements for buildings situated on our near landfill sites often do not include monitoring of CO_2 levels in the critical areas, but only of methane. It is however a well-known fact that methane can be rapidly oxidised to CO_2 in soils. Absence of monitoring for CO_2 will result in the LFG migration

going undetected, though it may still constitute a hazard (Damiani & Gandolla, 1992; Gandolla *et al.*, 1997).

3.2 Methane (CH$_4$)

Methane is the second main component of MSW landfill gas generated during the anaerobic degradation of the waste, where its concentration typically ranges between 30 and 60% of the gas under stable methanogenic conditions. Methane emissions from MSW landfills last for decades. They can be reduced by so-called mechanical-biological pretreatment of MSW which reduces the organic content of the waste before it is landfilled. Conversely, the start of methanogenesis and thus methane emissions may be retarded due to excessive accumulation of acids, insufficient or excessive moisture. It is also possible to speed up the initiation of the methane generation stage and increase the methane production rate by various operational measures, which help avoid the initial accumulation of acids or improve the water balance and distribution. In this way the duration of emissions may be reduced, but the total amount of emissions is not thought to be affected (Lagerkvist *et al.*, 1999).

Methane may also result from the anaerobic degradation of the organic carbon fraction in slag and ash or industrial wastes, such as sludges from the paper industry.

Methane is an important greenhouse gas. Despite its relatively low atmospheric concentration compared to carbon dioxide (currently 1.75 ppm as opposed to 358 ppm), the higher infrared absorption potential of methane makes its global warming potential some 27 times higher than that of CO_2 (mole for mole). The present global methane emissions are estimated at about 550 Tg yr^{-1} and the net annual increase at 40 Tg yr^{-1} (Dlugokencky & Masarie, 1998).

The total yearly methane emissions seem to have remained relatively constant since 1984, though the distribution of emissions may be changing and the emission rates from individual sources are not well enough defined to quantify trends in them, since the sources, both natural and anthropogenic, are all diffuse and thus difficult to quantify precisely (Conrad, 1996). Wetlands are the main natural source of methane, accounting for some 20% of the total emissions, about 110 ± 26 Tg yr^{-1} (Boeckx & Van Cleemput, 1996). In total, human activities presently account world-wide for some 70% of the emissions, with landfills as the third most important anthropogenic source. Various estimates have put the contribution of landfill CH_4 emissions at about 30-70 Tg of CH_4 yr^{-1}, an amount comparable to the current imbalance between sources and sinks (Boeckx & Van Cleemput, 1996).

Methane is generally not considered toxic to plants or other organisms. The major effect of methane in soils is believed to be due to methane oxidation which depletes the oxygen present, increases the carbon dioxide levels and may also raise the soil temperature. This may lead to plant death by enhanced asphyxiation due to a lower solubility of gases inside the plant cells or by drying up the soil.

The main hazard of methane for people and property are due to its flammable and explosive properties. The flammability range for methane in air at atmospheric pressure and ambient temperature is 5 to 15% and the safety concentration limit for confined environments where people live or work is 1% in air (20% of the Lower Explosivity Limit). Flammable mixtures will be explosive when contained in a small volume from which the products of combustion cannot readily escape. The potential of methane for flammability or explosion will also be influenced by the other components in the mixture. Thus no flammable mixtures exist when the diluent is carbon dioxide or nitrogen and the oxygen level falls below 12.8%. The flammability range is lower than the normal concentrations of methane in pure landfill gas. However, during migration, landfill gas will mix with air and become depleted in methane, through dilution and/or oxidation, and therefore fall within the flammability range. This is typically the case when methane migrates into a building through cracks in the foundations or through service mains. Dozens of reports of accidents involving the explosion or ignition of methane from landfill gas have been reported in recent years (Christensen, *et al.* eds, 1996; Gendebien *et al.*, 1992).

3.3 Hydrogen (H$_2$)

Hydrogen is a non poisonous, odourless and colourless, but highly inflammable gas. In MSW landfills, it is produced both by the fermentative and the acetogenic bacteria and consumed by the methanogens. As this last group of microorganisms are the slowest to develop in the metabolic succession of anaerobic degradation, molecular hydrogen may accumulate during the initial stages of waste degradation and be transiently present at high levels (up to 20%) in the gas phase of young landfills, well above its Lower Explosivity Limit of 4%.

Hydrogen is also produced in ash and slag deposits: elemental aluminium, which is the fifth most abundant element in bottom ash, reacts with water to form aluminium hydroxyde and gaseous hydrogen (Chandler *et al.,* 1997). Other metallic components with valence states of zero, such as zinc or copper, could also promote similar redox reactions resulting in hydrogen evolution. Heyer and Stegmann (1997) observed hydrogen evolution in simulated bottom ash deposits, with a maximum of 15%-vol right after deposition, followed by a steady decrease to zero over the next 40 days. No quantitative data on hydrogen evolution is however available. A variety of industrial wastes may have a potential of hydrogen formation on wetting.

No reports exist of accidents involving hydrogen on landfills. It is relatively easily oxidized in soils by micro-organisms and this has been documented experimentally in landfill top covers (Nozhevnikova et al., 1993).

3.4 Non methane organic compounds (NMOC's)

The gas from MSW landfills usually contains a variety of trace organics which may make up to 1%vol of the gas. The many published research studies have generally detected between 100 and 200 different compounds at each single site investigated (see Gendebien *et al.,* 1992, for a review). These trace components are either the sub-products of the biological and chemical processes occurring in the waste, and consist for example of oxidised carbon compounds: alcohols, ketones, esters, organic acids, furans and sulphur compounds. Or they may be of anthropogenic origin (such as halogenated hydrocarbons) and have been deposited together with the wastes. They will undergo various processes in the waste body and cover soil of the landfill, such as volatilisation, adsorption or biodegradation, which will govern their fate inside the landfill and their release into the atmosphere.

The concentrations of these compounds are extremely variable from one site to another and it is difficult to give typical values for specific landfill types. The nature of the wastes deposited and the rates and mechanisms of degradation occurring will have a decisive influence on the types of compounds and concentrations detected, as will the atmospheric conditions and the sampling and analytical procedures used. It is also often unclear whether the samples analyzed were recovered from deep in the landfill or at the effective emission sites on the landfill surface.

The distribution of these compounds between the solid, liquid and gas phases in the landfill may also vary widely from one compound to another. Their presence in the gas phase will depend on their polarity: more polar compounds will tend to solubilize in the leachate, while the more apolar ones will be found in a higher proportion in the gas phase. However, most NMOC's are sorbed for more than 98% to the solid phase, from which they will only be removed very slowly. Exceptions are vinyl chloride and some CFC's that one can expect to be 90% removed in less than five years (if one excludes any *de novo* synthesis from degradation of higher chlorination compounds).

The majority of the organic carbon in bottom ash is not characterised at this point in time, but it is thought to be mainly composed of unburned municipal solid waste (Chandler et al., 1997). Compared to the information available on the inorganic characteristics of bottom ash, the information on trace organics is very limited. Trace organics identified in bottom ash and which might be of potential concern for human health include polychlorinated dibenzo-p-dioxins (PCDD's), polychlorinated dibenzo-p-furans (PCDF's) and polyaromatic hydrocarbons (PAH's). The formation and partitioning of these compounds during incineration will greatly depend on the concentration of compounds or precursors in the waste, the type of incinerator and operating conditions. Improved process technology and control has resulted in the last years in a decrease of the presence of these compounds both in the flue gas and in the solid residues of incineration. Dioxins, furans and PAH's are semi-volatile organic compounds, characterized by a boiling point above 100°C, so their subsequent atmospheric emission from ash and slag deposits will in all probability be very slow. The heating induced by exothermic reactions occurring in such deposits, such as the

formation of metal hydroxydes (see paragraph 0), may however increase the atmospheric emissions of these compounds. No or only very little data is available.

Trace compounds generated during refuse degradation are generally themselves easily biodegradable and are associated with high degradation activities, *i.e.* younger landfills. They do not generally constitute a health or environmental hazard. Along with the sulphur compounds (see Table 3.1 and paragraph 3.5) they may however contribute to the foul smell of the gas.

Aromatic hydrocarbons, owing to their widespread utilisation, are widely detected in landfill gas (see Table 3.1). Compared to the emissions from other sources, such as traffic or chemical industries, emissions of aromatic hydrocarbons from landfills are certainly negligible with regard to their environmental effect. Nevertheless, some of these components, and in particular benzene, a proven carcinogen, may have adverse effects on landfill workers and should therefore be monitored. Biochemical degradation of aromatic hydrocarbons is generally very limited under methanogenic conditions, so they will mostly persist in the landfill.

Chlorinated hydrocarbons and other halogenated hydrocarbons will occur in MSW landfill gas due to contamination of household waste with solvents, propellants from spray cans and cleansing agents (see Table 3.1). They may also originate from wastes with a high plastic content, such as car-scrapping residues, which are known to increase the levels of adsorbable organically bound halogens (AOX's) in the landfill gas and leachate. The toxicity of the majority of these substances is rather low, but their persistence in the environment is generally very long, so their concentration in LFG has been extensively investigated (see Rettenberger & Stegmann, 1996, for a review). Biochemical degradation of halogenated hydrocarbons generates readily volatile non-polar substances, which will appear in the landfill gas. In some cases, it is the product of degradation of the original contaminants which present the greatest toxic risk. This is in particular the case for tricholoroethylene and the carcinogenic vinyl chloride, which result from the anaerobic biodegradation of tetrachloroethylene. Contamination of landfill gas with halogenated hydrocarbons may also cause corrosion problems in gas motors, due to acid formation upon combustion of the gas.

Table 3.1 shows that several anthropogenic NMOC's have been detected in landfill gas at concentrations exceeding the exposure limits. It should be noted however that most of the values cited for these compounds correspond to undiluted biogas, while the actual exposure levels are probably lower. Very few studies on the impacts of these compounds on the health of landfill workers have been carried out to date (see Loth, 1998 for a review).

Toxic or environmentally significant NMOC's are also emitted during landfill fires. A Finnish study (Ruokojärvi *et al*., 1997) showed that between 1990 and 1992, some 380 fires occurred annually at Finnish landfill sites, for a total of some 630 landfills. In the course of this study

two landfill fires, one experimental and one spontaneous, were also monitored and air samples collected. Unsurprisingly, PCDD and PCDF concentrations exceeded by some 4 orders of magnitude the limit value of 0.1 ng(I-TE) m^{-3} for emissions from waste incineration plants (see Table 3.1). It also appeared that the maximum acceptable daily intake values for these compounds would be exceeded for the fire-fighters and landfill staff, if pressurised breathing equipment was not used. The PCB levels measured during the two fires which were monitored did not exceed the NGV levels, and were related to the type of waste, since the fire involving industrial waste showed the highest levels of PCB's (Table 3.1). PAH's were also measured in the air near the fire (Table 3.1). A study conducted on Swedish landfills showed that the amounts of dioxins emitted annually from landfill fires might exceed by 3 to 4 times the amount emitted by the existing Swedish waste incineration plants (Pettersson *et al.*, 1996).

Some reports exist on emissions of PCDD's, PCDF´s and PAH's after combustion of LFG in flares, muffle furnaces or engines. PAH's result from incomplete combustion and are therefore formed primarily in case of operational problems. For dioxins and furans, *de novo* synthesis could also occur in zones of lower temperature of the combustion systems (> 250°C), but there is as yet little evidence to support this.

Little is known about the mitigation of LFG trace compounds in landfill cover soils. Methane oxidizing bacteria are known to be able to co-metabolize halogenated hydrocarbons, in particular tricholoroethylene (Hanson & Hanson, 1996). Preliminary field measurements on US and Danish landfills (Bogner et al., 1997, Kjeldsen et al., 1996) indicate that biodegradation or adsorption do occur in the cover soil and could substantially contribute to the mitigation of such emissions. Some of the trace components of LFG could also have an effect on the vegetation of the cover soil. Ethylene in particular, which has been detected in LFG at concentrations up to 180 ppm, interferes with the hormonal system regulating development and may have an adverse effect on plant growth (Maurice, 1998).

Table 3.1. Concentrations detected in MSW landfill gas for some aromatic and chlorinated hydrocarbons and sulphur compounds, and Swedish threshold limit values for exposure. A1, carcinogen, A2, induces mutations (Compiled from various sources: Christensen *et al.* eds, 1996; Gendebien *et al.*, 1992; Ruokojärvi, *et al.*, 1997; AFS, 1996).

Substance	Range reported (mg m^{-3})	NGV[a] (mg m^{-3})	KTV[b] (mg m^{-3})	Comment
Aromatic compounds				
Benzene	0.03 - 15	1.5	9	A1
Toluene	0.2 - 615	200	400	
Ethylbenzene	0.5 - 236	200	450	
Xylene	0.2 - 383	200	450	
Propylbenzene	1.5 - 173	-	-	
	24			
PAH's	810 - 1670[c]	-	-	
Chlorinated hydrocarbons				
Dichlorodifluoromethane (CFC 12)	4 - 700	2 500	4 000	
Trichlorofluoromethane (CFC 11)	0.4 - 500	3 000	4 500	
Chlorodifluoromethane (CFC 22)	2 - 276	1 800	2 500	
Dichloromethane	0 - 684	20	40	A1
Trichloroethylene	0 - 312	50	140	A1
Tetrachloroethylene	0.1 - 250	70	170	A1
Vinyl chloride	0 - 264	2.5	13	A1
Dichloroethylene	0 - 294	4	20	A1
Chloroform	0 - 50	10	25	A1

PCDD's / F's (pg(I-TE) m^{-3})	not detected 51 - 427[c]	5 pg(TE-nord.) kg^{-1}			100[d]
PCB's (ng m^{-3})	0.2 11 - 590[c]	10 000		30 000	A1
Sulphur compounds					
Hydrogen sulphide	0 - 20 000	14	20		
Methyl mercaptan	0.1 - 430	1 ppm	-		
Ethyl mercaptan	0 - 120	1 ppm	-		
Carbon disulphide	<0.5 - 22	16	25	A2	

[a]   Nivågränsvärde: threshold limit value per working day

[b]   Korttidsgränsvärde: threshold limit value (time-weighted average) for short exposure time

[c]   Baseline concentration in landfill air and concentration during a fire (Ruokojärvi et al., 1997)

[d]   Limit value for emission from waste incinerators in many European countries

[e]   NGV for sum of dimethyl disulphide, ethyl mercaptan and methyl mercaptan

The analysis of anthropogenic trace components in LFG gas may also be used for source identification in cases of suspected gas migration, to distinguish landfill-derived gas from other sources. In a study on a landfill gas plume emanating from a U.K. landfill, Ward and his co-workers (1996) detected 79 different volatile organic compounds, similar to those found in other landfills. The concentration profiles of some of the halogenated compounds suggested that they had been flushed out of the landfill during its early stages and persisted in the plume, since their concentrations outside the landfill were higher than the maximum levels detected on the landfill, even exceeding the NGV in the case of vinyl chloride.

3.5 Hydrogen sulphide (H$_2$S) and organosulphur compounds

Organosulphur compounds, *i.e.* mercaptans and carbon or methyl sulphides, are important contributors to the foul smell of MSW landfill gas (see Table 3.1). They most probably arise from the degradation of proteins, which typically form some 6% of food wastes. After an initial peak in concentration just after waste deposition, when the most easily degradable substrates are being consumed, their level in LFG has been shown to decrease rapidly (Gendebien *et al.,* 1992), to levels around 0.1 mg m^{-3} (3 ppm).

Generally, the levels of sulphur compounds are lower in LFG than in other biogases, below 100 mg m^{-3}, since the latter often contain a higher initial proteic fraction. In MSW landfills, volatile sulphur levels may be further decreased by formation of insoluble metal sulphides. However, if wastes containing even small amounts of organic carbon, or another reductant such as iron, are mixed with sulphate-containing waste, typically gypsum from C & D wastes, sulphide production can become important, in the range of 0.1 to 6 g m^{-3}, even up to 20 g m^{-3} or 59%vol (Rettenberger, 1996).

Sulphur is abundant in bottom ash and has been found in concentrations ranging from 1 000 to 5 000 mg kg^{-1} (Chandler *et al.,* 1997). Most of the sulphur is in the form of sulphate. Sulphur, in the form or sulphate and sulphite, is one of the major constituents of APC residues, with concentrations exceeding 10 g kg^{-1} (Chandler *et al.,* 1997). Since bottom ash and APC residues also contains small amounts of organic carbon, it is not possible to exclude microbial H_2S generation from slag and ash deposits.

Hydrogen sulphide is highly toxic and affects the nervous system. It also has a repugnant odour and is highly flammable. Its odour threshold is comprised between 5 and 40 ppm. Above 50 ppm it paralyses the olfactory system, which makes it a particularly pernicious intoxicant. Concentrations above 400 ppm affect the nervous system and above 700 ppm there is risk of death by respiratory failure. The NGV is 14 mg m^{-3} (10 ppm) and the KTV is 20 mg m^{-3} (15 ppm).

The most toxic of organosulphur compounds is methyl mercaptan, which affects the central nervous system. The NGV for total organic sulphides (methyl mercaptan, ethyl mercaptan and dimethyl disulphide) is 1 ppm. Concentrations well above this threshold, up to 430 mg m^{-3}, have been measured in landfill gas (Rettenberger & Stegmann, 1996).

H_2S and organosulphur compounds may well represent the most critical group of LFG components encountered to date, due to their toxicity, odour potential and corrosiveness. When in contact with water, H_2S gives rise to sulphuric acid, which may corrode gas utilization facilities. It may also result in high SO_2 emissions after LFG combustion. H_2S may be removed from LFG before its utilization by various processes such as oxidative washing, adsorption catalysis, chemical reaction and oxidation or passage through a biofilter or bioscrubber (Rettenberger, 1996).

3.6 Ammonia and nitrogen compounds

Ammonia and organic compounds containing nitrogen mainly methylated amines may also be found in MSW landfill gas. Like the organically bound sulphur they arise from the degradation of proteins. NH_3 concentrations up to 100 mg m^{-3} are recorded in the literature

(Christensen *et al.*, 1996), while little data is available for organically bound nitrogen. The NVG for ammonia is 18 mg m^{-3} (25 ppm), and its odour threshold is 1 ppm. Landfill workers could thus be exposed to above-limit levels, though the very low odour threshold of ammonia can be considered a security factor. Ammonia may also be released during leachate treatment, for example during aeration. Ammonia stripping is in effect still widely used as a way of reducing the nitrogen load of the leachate. The amount of ammonia stripped will increase with the pH of the leachate since the $NH_4^+ \leftrightarrow NH_3 + H^+$ equilibrium will be shifted towards NH_3 under alkaline conditions ($pK_a = 9.3$), which is the case for leachates from methanogenic landfills.

Some types of industrial wastes have a potential for higher ammonia emissions. For example, Bergman (1996) observed up to about 20%-vol of ammonia in the headspace of physical landfill simulators with spent potlining from aluminium production. The gas was generated on wetting of the wastes. Wastes generated at elevated temperatures and in contact with air may be expected to contain various volatile nitrogen compounds such as cyanides and ammonia.

Oxidised forms of nitrogen are unwanted products of the combustion of landfill gas and are largely influenced by the combustion technology and by parameters such as temperature, reaction time and oxygen supply. NO_x emissions are generated at temperatures above 1 000°C and reach their maximum at 2 000°C.

Nitrous oxide (N_2O) and nitric oxide (NO) may also be produced in soils during the microbial oxidation of ammonium. N_2O emissions in particular are environmentally relevant as it is a greenhouse gas with a global warming potential 300 times that of carbon dioxide. Of the Swedish contribution to the greenhouse effect, about 9% comes from N_2O. Observations on Swedish landfills (Börjesson & Svensson, 1993) have shown that N_2O emissions are influenced by the methane content of the soil, with concentrations above 5% stimulating N_2O formation. However, the microbiological background of this process is still poorly understood and deserves further study (see also chapter 0).

3.7 Carbon monoxide (CO)

Carbon monoxide has been found in municipal solid waste LFG at concentrations between 0 and 3%vol, with normal levels being around 0.001%vol (Gendebien et al., 1992). If higher concentrations are present, this may be a sign of oxygen-starved burning of the refuse. During an experimental landfill fire in Finland, Ettala et al. (Ettala *et al.,* 1996) measured up to 1600 ppm of CO inside the burning waste mass. The levels were highest during the extinguishing of the fire. CO may also be emitted during combustion of LFG, as a result of incomplete combustion. It is a highly toxic gas which binds to hemoglobin, thus causing respiratory failure and death above 5 000 ppm. Its NGV is 40 mg m^{-3} (35 ppm), and KTV 120 mg m^{-3} (120 ppm). Carbon monoxide can be oxidised by bacteria to CO_2 and this has been shown to occur in landfill top covers (Nozhevnikova *et al.,* 1993).

3.8 Mercury (Hg)

Municipal solid waste has been identified as a potentially significant source of mercury. MSW contains mercury due to the disposal of a variety of products which contain this metal. Mercury is found in varying amounts in batteries, fluorescent and high intensity light bulbs, thermometers, thermostats, light switches, with batteries as the most important contributor. A reduction of the contamination of MSW with mercury essentially depends on upstream measures, such as the reduction of the mercury content of batteries, which is the case in Sweden, and increased separate collection of household hazardous wastes.

During incineration, most of the mercury passes into the gas phase and from there into the APC residues, which concentrate 70 to 80% of the mercury input (Chandler *et al.,* 1997). Swedish incinerator slag contains an average of 0.20 mg kg^{-1} (Chandler *et al.,* 1997), which is a rather low value, only about a factor 10 higher than the average world-wide soil concentration.

Once released to the environment, mercury is distributed to the earth's surface including soils, wetlands, lakes, and oceans. It can then undergo chemical transformations, predominantly oxidation-reduction and methylation-demethylation. Biological processes play an important part in these transformations. Methylation, the addition of (-CH$_3$) to ionized mercury has been documented in water, sediments and soil. It is a microbial process and is in particular affected by dissolved oxygen, sulphur, organic matter or clay, and by the pH. More methylated mercury is formed in oxygen-limited environments and sulphate-dependant bacteria are thought to be involved in the process. These conditions are characteristic of MSW landfills, so it is probable that methylation may also occur there. Clay particles and organic matter bind the mercury, thus decreasing its bioavailibilty for methylation. It is in its methylated form that mercury bioaccumulates in higher organisms. Methyl mercury is toxic and fairly soluble in water. Dimethyl mercury is much less soluble, so it is less toxic. In anaerobic environments mercury may also be converted into the insoluble mercury sulphide.

Most information available on mercury in landfills concerns the solid and liquid phases. Elemental mercury has however been detected (10-24 ng m^{3}) in the air above Swedish landfills (Wallin, 1989). The concentrations were positively correlated to the air temperature. These levels, though significant in a wider environmental perspective, are 3 orders of magnitude lower than the Swedish NGV for mercury (0.03 mg m^{3}) and seem to indicate that mercury does not constitute an occupational health hazards in landfills.

Other elements may also be methylated, such as for example arsenic and platinum. At present there is no information available about such compounds in landfill gas.

3.9 Silicon (Si) compounds

Siloxanes, or silicones, are commercially produced compounds containing carbon, silicon, oxygen and hydrogen, and must be distinguished from inorganic silicon which contains no carbon, but only SiO_2 units. When combusted however, the product will be inorganic, regardless of its original form and the impact is believed to be the same.

The increasing utilization of landfill gas in reciprocating engines or turbines for energy recovery has sparked awareness of technical problems due to corrosion and fouling from residues generated by the combustion of trace impurities in the gas. The discovery of high levels of silicon in such deposits, combined with the volatility of specific siloxanes, has prompted investigators to name siloxanes from discarded commercial and consumer products as the primary source of these deposits and a major contributor to reduced engine lifetime (Niemann *et al.,* 1997). This has lead several engine manufacturers to add elemental Si limits on their equipment.

The identification of the sources of Si deposits in gas utilization equipment is complicated by several factors. Firstly, a large variety of siloxane compounds exist, due to their polymeric nature. Not only the low molecular weight volatile siloxanes are entrained, but high molecular weight siloxanes have been shown to aerosolize and thus be transported in gas streams. This variety complicates their quantification in the gas stream. Poly (siloxanes) are solvent soluble, but their degradation products, silanols, which known to occur in soils and may well also be present in landfills, are water soluble and may enter the LFG both by direct volatilization or through water entrainement. Secondly, naturally occurring silicon compounds are very common in landfill environments. Though they are exclusively non volatile mineral species, they may be transported with the gas as particulate matter or as solutes in water droplets.

Elemental characterization of combustion deposits in four US landfills and of Si compounds in the LFG gas and gas condensate (Niemann *et al.*, 1997) has shown that engine operational history (*i.e.* valve maintenance) is not only correlated to siloxane levels in the gas, but may also be influenced by other factors, such as the oil additives and the presence of acidic compounds.

3.10 Particulate emissions and aerosols

The emissions of dust and aerosols on landfill sites is of concern, due to their possible impacts on workers health. Some studies of dust levels on landfills (reviewed in Loth, 1998) showed that atmospheric concentrations above exposure limits may easily be reached, in particular inside the cabins of the compactors or of the loaders handling wastes or cover soil. The increase of waste sorting and pre-treatment activities carried out at landfill sites may also influence these emissions and consequently the exposure of the workers, though almost no data is at present available and this should be the object of further study.

3.11 Conclusions

The diversity of gas-phase compounds emitted by landfills is considerable. The impacts of the quantitatively most important emissions from landfills, methane and carbon dioxide, are well recognised today. Much research concerning these compound focuses on questions related to the global budgets of these two greenhouse gases. These aspects will be further discussed in chapter 0.

Generally speaking, information about their present levels of emission in landfill gas is lacking for most compounds outside methane. Furthermore, quantitative and qualitative predictions of their emissions are rendered difficult by the considerable uncertainties surrounding waste generation, in particular those aspects linked to public awareness and environmentally-friendly behaviour, *i.e.* the choice of "green-label" products, or the separation of toxic household wastes from the urban waste stream.

Present trends in legislation are also to incite the producers of the goods to offer more sustainable products, the elimination of which poses less or no threat to the environment. Therefore, in the long term, one should be able to expect a decrease of harmful emissions from landfills. However, many of the substances or compounds in question, in particular the NMOC's, are characterised by their persistence, and will continue to be emitted at low levels long after their elimination from the consumer goods. Table 3.2 summarises the impacts of the compounds discussed in paragraphs 0 to 0 and suggests possible future trends.

Some of these emissions will however simply be displaced towards the waste sorting and pre-treatment stages which are becoming more common. Green-waste treatment facilities and waste sorting plants are known to release large amounts of NMOC's and nitrous and sulphur compounds, which are often malodorous, as well as dust, spores, microorganisms and aerosols, which may adversely affect the health of the people employed there. As for landfills sites they are increasingly diversifying their waste treatment activities, and serving more and more as "multifunctional platforms", where different waste fractions are sorted, sent to the recycling industry, or pre-processed before landfilling.

These changes in waste management practice have not always, by far, been accompanied by an assessment of the atmospheric emissions and their potential ecological or occupational health impacts. Studies of the health impact of gas-phase emissions from landfills are at present extremely few (Loth, 1998), and mainly concern hazardous waste sites and therefore principally address the exposure to anthropogenic organic compounds or heavy metals. Sulphur compounds, despite their odour potential, high toxicity and the very low threshold limit values allowed for them, have not been studied. Some assessments have been made of the health effects and odour emissions from waste sorting and composting plants. Global comparative occupational health studies of various alternative treatment schemes are however non existent.

Similarly, few assessments have been made of the odours that may be generated by the expansion of such practices. Though their actual hazard potential is very low, foul-smelling emissions are possibly among the most deleterious in terms of image of a waste-treatment plant and their avoidance figures among the operators top priorities. Some assessments should therefore be made of the impacts that the new waste handling practices may have in terms of odours, as well as of measures that may be taken to prevent their development.

The trends that are now apparent in most European countries towards increasing separation of the putrescible fraction of waste and its treatment by composting or anaerobic digestion, as well as towards generalised incineration of waste fractions with high calorific value, will in the long-term, reduce the amounts of methane generated and emitted, but not those of carbon dioxide, which will rather increase, due to the increased use of aerobic processes.

The biochemical processes occurring in future landfills will probably change markedly over the next 20 years. Landfills are expected to receive proportionally more waste fractions which are not biologically active. However, emissions can occur from non biodegradable residues, such as AOX from car-scrapping wastes. Some consideration in particular should be given to the emissions from incinerator residues landfills, about which very little is known at present. The same applies for residues from waste sorting activities, such as for example scrapped tyres. In view of the reduction of the putrescible fraction of wastes going to landfills, while no or little change is expected for C & D wastes, the conditions favouring the emissions of hydrogen sulphide should also be investigated in more detail.

Despite these important changes in the composition of landfilled waste, the amounts of biogas emitted will still continue to be high for many years after the landfilling of large amounts of biodegradable waste has ceased and will in part also simply be transferred to anaerobic digestion plants. Investigations on alternative uses for this combustible gas, that are both ecologically and economically sustainable, especially for small landfills, should be encouraged.

With the economic sustainability of such plants in mind, the trace contaminants inducing corrosion and wearing in the gas utilisation facilities deserve further investigation. This is in particular the case for silicon compounds, for which the analytical characterisation and quantification in landfill gas is still problematic, so little is as yet known of their effective impact. A better knowledge as to their origins and fate, and the development of technical means of eliminating them from the gas are certainly to be hoped for.

Table 3.2. Impacts, relevance and trends of gas-phase emissions from landfills

Compound	Concentration range	Scale of the impacts	Type of impacts	Relevance	Expected trend of emissions
Carbon dioxide	20-50%	global	global warming	high	↗
		local	vegetation, asphyxia	high	
Methane	30-60%	global	global warming	high	↘
		local	vegetation, asphyxia	high	(slow)
		local	explosion and fire hazard	high	
Aromatic HC's	mg m^{-3}	local	health hazard	low	↘
Halogenated HC's	mg m^{-3}	global	environmental persistence	high	↘
		local	health hazard, corrosion	high	
CFC's	mg m^{-3}	global	global warming	low	↘
PCDD / F's	mg m^{-3}	local	health hazard	punctually high (fires)	→
PAH's	mg m^{-3}	local	health hazard	punctually high (fires)	→
Other NMOC's	mg m^{-3}	local	odours, vegetation (ethylene)	low	↘ (slow)
Hydrogen sulphide	0 - 20 g m^{-3}	local	health hazard, odours, corrosion	high	→

Organosulphides	mg m^{-3}	local	health hazard,	high	→
			odours, corrosion	low	
Carbon monoxide	0 - 3%	local	asphyxia, explosion	low	↘
Hydrogen	0 - 20%	local	explosion	low	→
Mercury	10 - 24 ng m$^{-3\ a}$	global	dispersion and bioaccumulation	unknown	↘
		local	health hazard	unknown	
Si compounds	mg m^{-3}	local	wearing of equipment	unknown	↗
Ammonia	mg m^{-3}	global	NOx formation	low	↘
		local	odours, health hazard	low	
Nitrous oxide	mg m^{-3}	global	global warming	unknown	possibly ↗
Nitric oxide	mg m^{-3}	global	acid deposition	low	↘
Dust / aerosols	mg m^{-3}	local	health hazard	high for workers	possibly ↗

[a]   Concentration measured in the air above Swedish landfills

4. Landfill processes and gas emissions

4.1 Gas balance and processes governing emissions

All gases formed within a landfill will eventually leave it, driven either by differential pressure or by concentration gradients. Gas transport processes include both spontaneous gas exchange over the landfill perimeter and active gas abstraction as indicated in Figure 4.1. In a properly managed landfill, lateral gas migration should be equal to zero, but this is not the case in reality. If it is different from zero, it could also be included in the "emission" term, since in the long run, the gas migrating in the sub-soil will also be emitted to the atmosphere, with or without conversion (*i.e.* oxidation). Transport by leachate and storage inside the landfill should also be included in this balance and could be of some relevance, especially for gases with a high solubility in water, such as carbon dioxide.

Carbon dioxide emission

Carbon dioxide and methane emission

LFG recovery or combustion

Carbon dioxide emission

Microbial methane oxidation

Methane and carbon dioxide storage

LFG generation

Carbon dioxide and methane migration

Carbon dioxide in leachate

Modified from Bogner & Scott, 1997
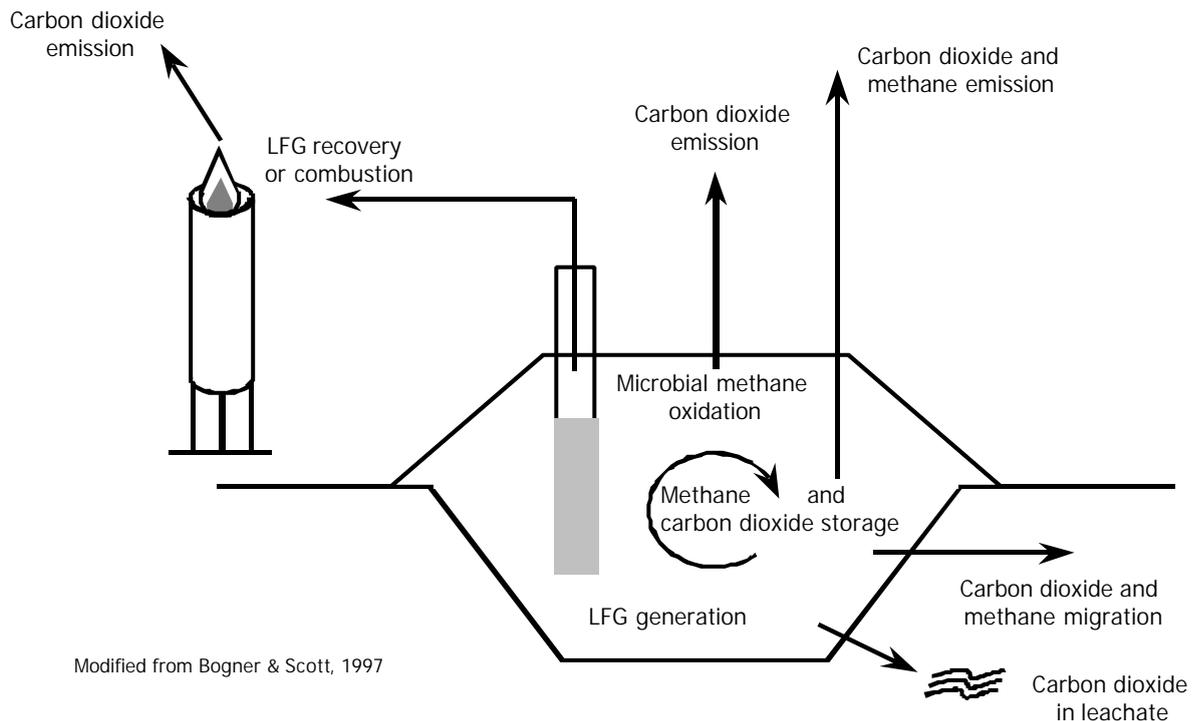
Methane generated =   methane abstracted + methane emitted +
                       methane migrated + methane oxidized + difference in methane storage

Carbon dioxide generated =   carbon dioxide abstracted + carbon dioxide from methane oxidation +
                              carbon dioxide from methane combustion + carbon dioxide migrated +
                              carbon dioxide in leachate + difference in carbon dioxide storage

Figure 4.1   Methane and carbon dioxide balance of a MSW landfill

Due to its importance as a greenhouse gas, methane has been by far the most studied atmospheric emission from landfills. Therefore, most of the methods, findings and open research questions we will be discussing in the following paragraphs concern methane emissions. However in the future, the emphasis may well shift somewhat to other compounds, as changes in waste composition and management practices will result in notable changes in the nature and level of landfill atmospheric emissions in the coming decades.

4.1.1 LFG generation

Gas formation in landfills depends on the wastes landfilled, the environmental conditions at and in the landfill and on landfill technology. LFG composition is the result of degradation processes in the landfill, as well as of evaporation of volatile substances and exchange of gaseous compounds between the landfill and the atmosphere. The composition of the gas will also in itself affect landfill processes, and thus the formation of gas. Oxygen intrusion, for example, may trigger aerobic degradation in parts of the landfill, and thus increase CO_2 production with respect to methane, but may also enhance the volatilisation of some compounds due to the elevated temperatures reached by aerobic degradation processes.

When predicting landfill gas formation, the focus is usually put only on biogas formation as a result of the anaerobic degradation of the biodegradable fraction of the waste, neglecting the fraction resulting from other processes. A potential gas generation can be calculated from the composition of the substrate. Empirical data is used or anaerobic degradation tests are carried out. The rate and ultimate yield of LFG is highly variable from site to site. A typical yield may be in the range of 200-300 $m^3\, t^{-1}$ of fresh MSW and the range of methane generation may range from below 1 m^3 up to about 40 m^3 per tonne and year (Lawson *et al.* 1992).

The models used for predicting LFG generation are based on the waste composition, *i.e.* its carbon content, degradability and the kinetics of degradation. Thus, other processes, such as volatilisation, are not taken into consideration. The most widely used model in Europe is probably Tabasaran's expression (Tabasaran 1976), a relationship originally developed for the anaerobic digestion of sewage:

$$G_a = G_e\, (1 - e^{-ka}),\qquad\qquad\qquad\qquad \text{(Eq. 3.1)}$$

where:

G_a: accumulated gas generation until year a [$Nm^3 t^{-1}$]

G_e: gas formation potential [$Nm^3 t^{-1}$]

k: degradation constant = $ln2/t_{\frac{1}{2}}$ [time unit^{-1}]

a: time [number of time units]

$$G_e = 1.868\ C_0\ (0.014T + 0.28)$$

        C_0:   content of degradable carbon in the waste [$kg\ t^{-1}$]

        T: temperature [°C]

The temperature correction in the G_e determination is derived from the fact that the portion of substrate which is used for cell synthesis may vary with temperature. Since the "sludge retention time" of landfills is on a completely different scale than that of an anaerobic digester, the correction is irrelevant for landfills.

In the USA, the Scoll Canyon model occupies a similar position to Tabasaran's model in Europe. This is also a first order decay model and it does not have the questionable temperature correction. For the same situation, both models give identical results. The USEPA used the Scoll Canyon model for the LANDGEM program which is published on the net (http://www.epa.gov/oar/oaqps/). This model can also be used to calculate emissions of a number of trace components, but based on fixed concentrations in the LFG, *i.e.* as a function of biogas generation.

A first order decay is the usual assumption in LFG generation modelling, but zero order models and higher order models have also been used. According to van Oonk *et al.* (1994), precision will increase with the order of the model. Furthermore, an additional refinement can be applied to models irrespective of their order, in the form of different degradation constants for different waste materials, so-called multi-phase modelling. However, waste composition data grouped according to degradability is then needed to allocate the different time constants. Contrary to van Oonk's results, a validation of gas generation models of zero, first and second order using operational data from 17 american landfills indicated that only small or no improvements could be gained by using higher order models (Augenstein *et al.* 1996).

If one considers the fact that reaching a similar degree of degradation takes decades in landfills and only days in anaerobic digestors (Bogner & Lagerkvist 1997), it seems likely that other factors than substrate availability may be equally important for gas

generation in landfills. Today, there is an information gap needing to be bridged about the impact of various factors on the degradation of landfilled waste. Although a number of factors are known to have an impact, the knowledge is in part only qualitative. In particular, very few studies have been undertaken where the incoming wastes have been characterised and the landfill conditions monitored with sufficient precision. The data available from model experiments is richer, but its interpretation is not always easy. One problem with model experiments is their temporal limitation. Since there will be a lag time for many biological reactions, some processes may stay undetected during such experiments.

Over the last 30 years, numerous studies aiming towards a better understanding and control of landfill processes have been undertaken. Over the last decade some larger-scale studies have focused on different methods to enhance LFG generation, while maintaining a good control of landfill gas emissions (Ecke & Lagerkvist 1997). Usual methods for improving gas yields include water addition, seeding (with sludge), heating and the addition of pH buffers and nutrients.

The first measure in order to achieve a better control of landfill processes is to control and select the waste materials entering the landfill or landfill part, since no techniques for enhancement of degradation or other control measures, are universally applicable to all waste materials. To allow for the separation of materials with different properties, a landfill needs to be divided into cells. Another good reason for this, is that it permits to reach filling height faster, and therefore also to seal the top faster.

Experiences of cellfills are relatively limited today, but it is already clear that landfilling in cells in itself promotes a better control of the processes occurring in the landfilled waste and above all, a more efficient capture of the gas formed. Simply through the disposal of degradable wastes in specially designed landfill cells, called biofills, the recovery of landfill gas can be increased 2 to 10-fold in comparison to conventional mixed landfills (Lagerkvist 1997). Other advantages of the biofill technique are lowered emissions of greenhouse gases, increased energy efficiency and an easier reclamation of the digestate in the future. The results of the co-ordinated Swedish LFG program (Lagerkvist 1997) indicated that methane emissions could be kept down to less than 10% of the methane formation. Cellfilling also leads to improved routines and better organised landfills, accompanied by volume savings and reduction of problems due to birds, wind littering, noise and visual disturbances.

4.1.2 Gas abstraction

Gas abstraction systems were originally developed from groundwater abstraction technology. The adaptation of this technology to landfill conditions and for landfill gas abstraction has been the object of much testing by waste companies and engineering consultants, but these investigations have rarely been systematically and properly documented. The know-how exists however, and it may be time for a compilation and

critical evaluation of present practices and opinions. For example, there is a widespread assumption that gas abstraction will affect gas generation, but the mechanisms by which this would occur are not clear. It has already been shown that the abstraction of LFG can be substantially improved by better structured landfills and by increasing the active surface of the gas collectors (Lagerkvist 1997), but there also exists a potential for improving the economy of gas abstraction through better designs and materials, especially with regard to maintenance.

Today gas is abstracted from landfills by distributing a partial vacuum from one or several sources through a grid of collection points. For a deep and large landfill with a thick cover, this is generally a workable solution, but there is a need to improve techniques for sites which are more sensitive to wind and other weather impacts. A better understanding of gas movements inside landfills would be useful for improving the design of collection systems. Instead of drawing on the experience of groundwater abstraction from uniform sediments, it might be possible to use models derived from the study of fractured bedrock for example, which may be more representative of the situation inside a landfill.

There is also a need to develop the automatic monitoring and control of landfill gas abstraction systems. There are a number of questions touching on the control of gas abstraction which are poorly elucidated today, stretching from which analytes should be used as inputs in a control system, over to which techniques could be employed to handle the mass of analytical data which is generated by such systems and exploit it for real-time data utilisation. Some promising results have been reported from the use of real-time multivariate analysis (Birkeland 1996) and this needs further research and development. Landfill gas monitoring is also coupled to the general problem of landfill information management – an issue which comes repeatedly to the fore, *e.g.* when a new environmental impact from landfills is discovered or suspected. Gas generation and emissions are in effect correlated to many, if not all, other aspects of landfill planning, operation and control, be it the waste types, amounts and handling, the leachate quality and management, the landfill topography and design. Doubtless much could be improved in the quality of the collected data and in its successive exploitation, by the development of geographic information systems (GIS) for landfill management, where data from various sources and of different types could be connected and analysed .

4.1.3 Emissions

Emissions vary widely over time, both diurnally and seasonally. Several environmental factors may influence both the gas quality and its flux, *e.g.* changes in barometric pressure, rainfall and temperature. In addition, during the transport of the gas through the soil cover many processes may occur, which will modify the composition of the gas phase, such as the adsorption and oxidation of organic compounds or their solubilization in the liquid phase.

An important conversion process is the microbial oxidation of methane. Like any process involving microorganisms, methane oxidation is sensitive to factors such as temperature, moisture and nutrient availibility. The soil type and structure has also been shown to influence methane oxidation and certain compounds are known to act as inhibitors. Ammonium and nitrogen fertilisation for example have been found to affect methane uptake, both in landfill and other environments (Conrad, 1996), and their effect has been much investigated. In the case of landfill environments, the trace components of the gas itself may actually also interact with methane oxidation processes and more attention should be given to this aspect, which is specific of landfills. Bogner *et al.* (1997a) observed that the levels of several NMOC´s were reduced during the transport of landfill gas through the cover soil, though the processes involved, whether microbial of physico-chemical, were not elucidated. Similarly, Kjeldsen *et al.* (1996) observed degradation of aromatic hydrocarbons and chlorinated aliphatics in landfill soils. Degradation rates were higher in the presence of methane, and, in the case of trichloroethylene, degradation only occurred when methane was present.

Methane oxidation in soils has been studied extensively in the laboratory, using mainly soil incubation experiments (batch tests) or soil microcosms flushed by a continuous gas flow (see Humer, 1997, for a review), but field-scale experiments are still few. In one case, data obtained from laboratory investigations was combined with field measurements of CH_4 emissions, to quantitatively estimate oxidation at a series of locations on a landfill and then extrapolate the data to the whole landfill (Czepiel *et al.*, 1996). Quantification of methane oxidation at field scale poses special challenges in the landfill setting. One important aspect to consider is the integration between active gas abstraction systems and methane oxidation in the cover soil, in particular the adaptative capacity of microbial population to variations in the methane flow. Landfills environments have been shown to harbour methane oxidizing populations adapted to different levels of methane and oxygen and to also, in some cases, act as sinks for atmospheric methane (Bogner *et al.*, 1997b). It is of interest that such soils harbour large and adaptable enough populations of microorganisms to take care of eventual emission peaks related to disturbances in the LFG collection system. The dynamics of methane oxidizing populations, as well as the conditions allowing for their maximum efficiency in landfill environments need therefore to be further studied.

Understanding the processes occurring depends first of all on the possibility of adequately measuring the amount and nature of the emissions. As we shall see in the following paragraph this is in itself not a simple problem, due to the complex dynamics and spatial variability of gas generation and movement in landfills.

4.2 Methods for measuring gas emissions

Gases move through the subsurface under the influence of both concentration and pressure gradients, that is both diffusion and convection. Usually one process is

dominant: diffusion may dominate in near-surface soils, while convection dominates near gas recovery wells, which are subject to partial vacuum in order to abstract the gas from the landfill (Bogner & Scott, 1997). Once they reach the atmosphere, their movement is dominated principally by convection and turbulent transfer processes.

No single perfect method exists to precisely quantify net emissions. However, choices are available between several techniques which have been tested and perfected in various field situations. Some are used to estimate emissions from small defined areas and thus give "punctual" LFG emissions, while others allow global estimation of the fluxes for larger areas, for example a whole landfill. Chamber techniques and subsurface vertical gradient measurements belong to the first group, while micrometeorological methods, isotopic fractionation, tracer techniques, Fourier Transform Infrared spectroscopy and remote observations belong to the second. In the following paragraphs we will comment on the use and characteristics of the most important of these methods. More detailed information is available in (Harper et al., 1993; Bogner & Scott, 1997; Heie & Lagerkvist, 1998).

4.2.1 Chambers techniques

Chamber measurements consider the gas concentration increase in a known volume of air, through a known soil surface area during a certain time. They have notable advantages, in particular their low cost and rapid data turn-around, their applicability to numerous types of gaseous emissions and the possibility of adjusting their sensitivity to site conditions by varying their volume. Their main disadvantages are that they are labour intensive and may affect conditions at the soil/atmosphere interface. However, this can be minimised by operating them over short time periods and avoiding disturbance of the soil surface. Chambers have been used extensively for measurements of methane fluxes on MSW landfills, but also to measure gaseous emissions in natural environments, from spill sites, contaminated land sites and hazardous waste deposits (see Bogner & Scott, 1997 for a review).

Chambers may be used statically or dynamically. A static chamber consists of a sealed air volume placed over the landfill surface, in which the variation in the concentration of the gas examined is measured over a short period of time, commonly between 30 min and a couple of hours (Figure 4.2.1a). During this time, the chamber atmosphere should be sampled at least 4 to 6 times (Bogner & Scott, 1997). Since the gas flow is caused by diffusion, the LFG concentration will reach a maximum in the static chamber and the net emission will decrease to zero. The change in concentration is plotted as a function of time and the slope and regression coefficient of a linear regression curve are calculated.

In dynamic chambers, a known air flow passes through the chambers (Figure 4.2.1b). The gas concentrations in the air are measured before and after the chamber and the

LFG emission rate is calculated from the variation in concentration and the flow. Dynamic chambers allow to avoid the effects of concentration build-up. Furthermore, the gas flux also ensures air mixing inside the chamber. But the inlet and outlet flows have to be measured with precision and differences in pressure between the different parts of the system may affect the flow pattern.

These techniques are particularly useful to obtain indications on the effect on gas fluxes of site-specific factors such as surface characteristics, soil moisture, vicinity of gas abstraction wells, and have been used extensively on landfills (Bogner & Scott, 1997; Bogner et al., 1997b; Börjesson & Svensson, 1997). To obtain the global fluxes for an entire landfill requires a large series of chamber measurements randomly distributed on the whole surface, such as described by Lagerkvist (1997).

4.2.2 Subsurface vertical concentration gradients

This method described by Rolston (1986) is based on Fick's law of diffusion. The soil-gas diffusivity and the gas gradient in the soil are needed.

$$F = - D_s \, (dc/dx) \qquad \qquad \text{(Eq. 4.1)}$$

Where:

D_s : soil-gas diffusivity ($m^2 \, s^{-1}$)

dc : vertical concentration difference for dx ($g \, m^{-3}$)

dx : vertical depth difference (m)

F : gas emission ($g \, m^{-2} \, s^{-1}$)

The soil-gas diffusion constant may be calculated or measured. The gas gradient is measured by inserting into the soil probes containing one or several cavities in contact with the soil atmosphere and from which the gas may be sampled (Figure 4.2.2)

Subsurface vertical concentration gradients have not been much described in the literature, but the technique is relatively simple and inexpensive. Just as for the chamber measurements, this method only gives punctual values and flux values will be strongly affected by variations in the soil cover or the presence of cracks. When compared with chamber measurements, slightly higher fluxes are often obtained. This may be caused by air intrusion into the upper soil as the gas samples are extracted. Fluxes may also be

overestimated if the gas analysed undergoes partial oxidation during its passage through the soil, as may often be the case for methane. This will result in a steeper gradient and therefore in an overestimate of the flux (Lagerkvist, 1997). Another disadvantage may be that the equation considers diffusion only. Gas pressure differences in the shallow subsurface are also a driving force influencing gas emissions (Bogner & Scott, 1997).

Micrometeorological methods

These methods are used to determine gas fluxes above the soil/atmosphere interface and can address broader time and space scales (Figure 4.2.3). They are based on the assumption that the gas fluxes measured in the lowest layer of the atmosphere, called the planetary boundary layer (PBL), represent the gas flux from the soil cover. This assumption is valid if the soil cover is uniform and the fluxes are steady with time. The thickness of the PBL varies diurnally due to surface heating and cooling processes, winds, surface topography and other factors. According to (Bogner & Scott, 1997), to be considered as uniform, a surface should extend for a distance 75 to 100 times the flux measurement height.

Above-ground gradient technique

The vertical flux of a trace gas is measured using the gas gradient in the atmosphere and the diffusion coefficient of the gas. The major difficulty is the estimation of the diffusion coefficient, varying for different gases (Bogner & Scott, 1997). The analysis equipment must have a high resolution, high precision and fast response. The topography of the surface investigated should be uniform and small-scale variations are not detectable (Schütz & Seiler, 1989).

Eddy correlation technique

The dominant mechanism for gas transport above ground is turbulent winds. Two parameters have to be measured, the vertical wind speed (W_v) and the trace gas concentration (C_g) over a sufficiently large interval at one height above the measuring site (Schütz & Seiler, 1989). The following equation gives the flux (F):

$$F = W_v \, C_g \hspace{6cm} \text{(Eq. 4.2)}$$

The main advantage of these methods is that they give information concerning a large area. They can be automated and are therefore useful for the study of variations over time (daily or seasonal variations, for example). But they require a sophisticated and expensive instrumentation, with a fast response (eg. sampling rates of 5 to 10 times per second) and suffer from surface constraints, requiring flat continuous landscapes (Bogner & Scott, 1997).

4.2.3 Tracer techniques and FTIR spectroscopy

These methods are based on the measurement of an inert tracer gas released at a known rate and the gas of interest. The concentration ratio of the two gases is measured at different locations around the landfill where a plume of the gas develops downwind (Figure 4.2.4). The gas emission rate from the landfill is calculated from the ratio measured in the plume and the volume of tracer released. Bogner & Scott (1997) reported that SF_6 has been used as tracer gas, mostly to study natural gas emissions from pipelines.

An alternative to SF_6 for tracer work has been tested by Galle and coworkers (1999) on Swedish landfills. N_2O was used as tracer gas and concentrations of N_2O and CH_4 in the dispersal plume were measured downwind from the landfill using LPFTIR (Long Path Fourier Transform Infrared) spectroscopy.

LPFTIR is an optical technique based on infrared absorption. Infrared light is transmitted over a certain distance through a gas and an absorption spectrum of the gas is recorded. Spectral analysis allows to determine the concentrations of different gases of interest (CH_4, CO, CO_2, N_2O, H_2O), simultaneously and with high time resolution. By the use of long optical path lengths, 50 - 1000 m, sensitivity of the order of a few ppb is obtained.

The same authors also used FTIR to measure the source distribution of methane emissions on the landfill by measuring CH_4 concentrations along parallel transects covering the landfill surface and applying a Gaussian dispersion model to calculate source strengths for each sector of the measurement grid.

Tracer methods and FTIR circumvent the problem of spatial heterogeneity of the source, by integrating the whole area flux. They seem therefore promising for estimating emissions from whole landfills. According to Galle *et al.* (1999), they should be further developed, as a cost effective and reliable way of inventorying total emissions from a large number of sites and of investigating their dependence on climatic factors such as wind speed or temperature. Disadvantages of these methods are their dependence on favourable weather conditions, the use of potent greenhouse gases as tracers and the potential for interfering sources of the gases investigated.

1 a)   b)

Inlet   Outlet

Clay sealing

Landfill surface

2

3   Measurement tower

LFG gradient

Landfill Surface

4   Main wind

Landfill

Tracer plume

Emission point of the tracer

Investigation transect

35
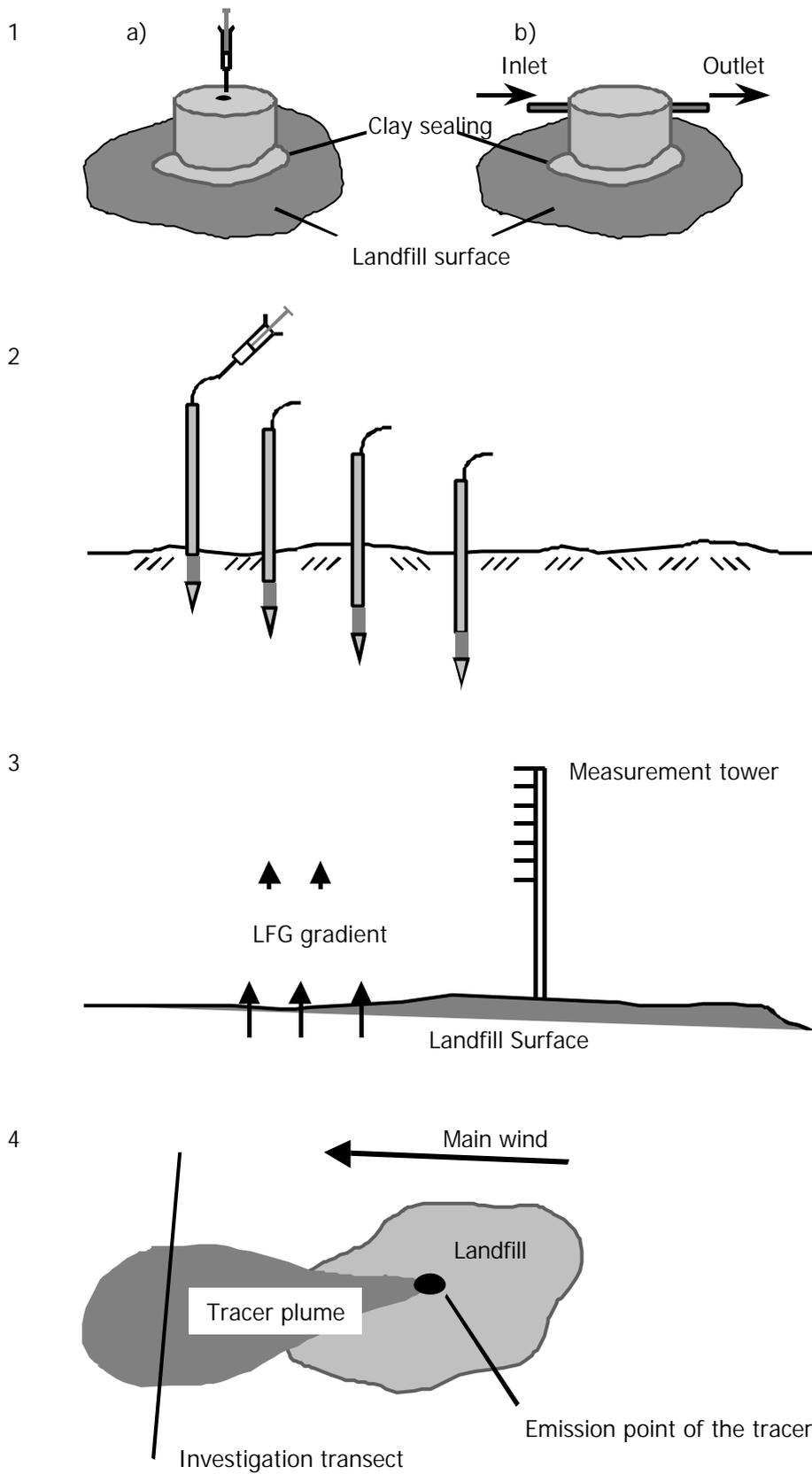
Figure 4.2.   Emission measurement techniques. 1 Chamber technique: a) static b) dynamic. 2: Subsurface concentration gradient. 3: Micrometeorological methods. 4: Tracer and plume dispersion.

4.2.4 Isotopic fractionation

When several isotopes exist for one element, as in the case of carbon, it has been shown that biological processes preferentially use the lighter isotopes. Thus the isotopic composition of methane varies depending on its origin. Methane resulting from microbial anaerobic degradation is enriched in ^{12}C and depleted in ^{13}C compared to methane produced thermogenically during the oil formation process. It has also been shown that bacteria oxidise ^{12}C more rapidly than ^{13}C. As a consequence, methane escaping from areas where oxidation occurs will be enriched in ^{13}C (Coleman *et al.,* 1981). Oil companies have used the technique to identify the origin of methane *e.g.* between methane from oil deposits and other sources.

Measuring the isotopic composition of a compound is therefore not *per se* a flux measurement. However attempts have been made to use stable carbon isotopes to understand the contribution of the various methane sources to the annual atmospheric burden (Levin *et al.*, 1993) and therefore obtain global values for atmospheric emissions on a national or even wider scale. However, all biogenic methane sources, including landfills, have overlapping base values of isotopic fractionation. Isotopic analysis of landfill methane seems at present to offer more promises for the elucidation of site-specific methanogenic and methane oxidation processes and determining the methane source in case of suspected migration (Ward *et al.*, 1996), than to assess global methane emissions from various sources.

4.2.5 Remote LFG emission measurements

Regional techniques address larger-scale landscape fluxes. Aircraft or satellites could be used. As landfills are of limited area, these methods have little applicability to direct measurements of landfill fluxes. In the case of airborne systems, they basically apply the eddy correlation method to a movable platform. (Bogner & Scott, 1997). Planetary boundary layer budgets are another method that may also be applied to larger area fluxes. In this technique, the inversion of temperature profile in the layer acts as a lid to define an upper boundary of a "box" on which the mass balance is carried out. These methods are not directly applicable to landfills for reasons of scale.

4.3 Modelling emissions

4.3.1 Methane

Methane is at present the most environmentally significant gaseous emission from landfills, since, alongside CO_2, it is quantitatively by far the most important, and furthermore contributes mole for mole more to global warming than CO_2. A better knowledge of the amounts and trends for each of the individual sources that contribute to the methane budget are urgently needed, so that the future evolution of atmospheric methane and its role in climate change may be predicted with the measure of accuracy required to pinpoint which sources could be targeted for reduction, most effectively and at the lowest possible cost.

In the following paragraphs we will therefore discuss in detail the problems related to the assessment and modelling of methane emissions, using a "top-down" approach, *i.e.* starting with the methodology used for the calculation of the contribution of landfills to national budgets and then discussing how small-scale emission measurements, better knowledge of processes governing gas generation and improved control of gas abstraction could help improve these estimates. Finally, we will also consider what other questions small-scale emission measurements can serve to address.

As opposed to natural methane-generating environments, such as wetlands, landfills represent potent quasi-punctual sources which are not readily modelled on a global scale, using climatic/vegetation parameters (as is for example the case for methane emissions from natural ecosystems). Landfills, where methane is produced during anaerobic degradation of organic matter, constitute approximately 20% of the anthropogenic sources. Of the Swedish contribution to the greenhouse effect, 78% comes from CO_2 , 8% from CH_4, 9% from N_2O and 5% from CFC's and SF_6 (www.environ.se). Methane originates mainly from the agricultural and waste treatment sectors, with the contribution of the waste sector estimated at 2% of the emissions of greenhouse gases in Sweden (www.environ.se). The annual methane emissions from landfills in Sweden are presently estimated to be 145 Mm^3 (RVF, 1996). All these figures, and all national and global budgets, are still based upon very general assumptions about the number of landfills world-wide and the methane generation and emission rates.

A methane balance provides a useful framework to describe landfill methane processes and net emissions at individual sites (Figure 4.1):

Methane production =

Σ (emission + lateral migration + recovery + oxidation + Δ storage) (Eq. 4.3)

For national (country-based) estimates which are summed to provide global estimates, a simplified methane balance is applied:

Methane production =

Σ (methane emission + methane recovery + methane oxidation) (Eq. 4.4)

In practice, equation (4.4) is re-written in terms of methane emissions and applied by the Intergovernmental Panel on Climate Change (IPCC) in its default methodology for calculating methane emissions from solid waste disposal (IPCC, 1996). The IPCC default method is widely used by countries throughout the world to estimate national methane emissions from solid waste disposal sites. Methane production for a given country is computed from waste generation data (either measured or calculated). The net methane emissions for a country are then estimated as follows, and national estimates are summed to provide a global estimate (IPCC, 1996):

Methane Emissions (Gg yr^{-1})

=

$(MSW_T \cdot MSW_F \cdot MCF \cdot DOC \cdot DOC_F \cdot F \cdot 16/12 - R) \cdot (1 - OX)$ (Eq. 4.5)

where:

MSW_T = total MSW generated (Gg yr^{-1})

MSW_F = fraction of MSW disposed to SWDSs

MCF = methane correction factor (fraction)

DOC = degradable organic carbon (fraction)

DOC_F = fraction DOC converted to landfill gas (dissimilated)

F = fraction of CH_4 in landfill gas (default is 0.5)

R = recovered CH_4 (Gg yr^{-1})

OX   = oxidation factor (default is 0, pending availability of new data)

This mass balance relies on several assumptions. First, all the potential methane is released the year the waste is disposed, *i.e.* the annual waste disposal rate is constant. Secondly, DOC ranges from 0.08 to 0.21 and the fraction of DOC dissimilated is 0.77 (default). The methane correction factor (MCF) ranges from 0.4 to 1.0 and accounts for the methane generation potential of the site which will depend for example upon the available oxygen, the level of compaction of the waste and the use of other. Finally, emphasizing the fact that this methodology estimates CH_4 generation rather than emission, and that oxidation often occurs in the upper layers of the waste mass and in site cover material, a CH_4 oxidation factor (OX) is included in the equation, but is currently set to 0 by default in the IPCC guidelines, pending the availability of further data.

In reality it is at present difficult, if not impossible, to develop a single national estimate for methane emissions given the current uncertainties in waste generation and composition, quantities landfilled, methane generation and net methane emissions. The only term in the budget which may at present be quantified with tolerable precision in many countries is gas recovery. In Sweden, biogas is extracted from more than 60 landfills, amounting to about 90 million Nm^3 in 1994 (RVF, 1996), for an energy utilization of 0.42 TWh.

Corrections to the IPCC model have been proposed by several authors. In many countries it is difficult to obtain data on waste generation. The IPCC model assumes that the waste generation is constant, *i.e.* if data is available for a given year for which methane emission estimations are being made, waste production in the preceding years is assumed to have been equivalent. Irving *et al.* (1998) showed that if one assumes instead that waste generation has been increasing at a constant rate, this may result in a more than 20% reduction of the methane emissions estimates for a specific country. Bogner & Matthews (1999) tested the applicability of using per capita energy consumption as a surrogate to waste generation. The correlation of this proxy to waste generation was linear, with a regression coefficient equal to 0.53 for developed countries and 0.96 for developing countries.

The other terms of the IPCC model are also difficult to estimate with precision. For example, the fraction of organic carbon dissimilated (DOC_f) is certainly much lower in non-optimised systems than the default value of 0.77 proposed by the IPCC model. The uncertainty about the DOC_f seems to lead to an overestimation of the methane production. In their recent study, Bogner & Matthews (1999) calculated global emissions using their surrogate for two scenarios: high rate of organic carbon conversion to methane and no methane oxidation ($DOC_f = 0.77$, OX = 0); low rate of organic carbon conversion and 10% methane oxidation ($DOC_f = 0.5$, OX = 0.1). The

value taken for the methane oxidation factor was based on the study by Czepiel *et al.* (1996) in which field data were used to estimate fractional oxidation of approximately 10% at a US landfill without gas recovery. The resulting estimates of net global methane emissions were in the same range as that obtained using the default IPCC methodology, but showed greater interannual variations, reflecting economic changes expected to result in altered waste generation rates.

More realistic figures for the amount of waste generated do not however suffice to improve the default methodology reliability, since the calculation of the methane generated from this waste is still based on very crude global assumptions regarding the methane generation potential of the waste and only includes a very general correction factor to account for a very wide range of landfills typologies and operational practices, to say nothing of general waste management strategies.

For example, the methane correction factor of the IPCC model, which is in effect designed to account for waste depth and level of landfill management, actually assigns a default value of 1 to all so-called "managed" landfills, *i.e.* those that practice at least one of the following measures: use of cover material, mechanical compacting or levelling of the waste (IPCC, 1996). Thus, all Swedish landfills should be given assigned an MCF of 1, while as we have seen in chapter 2 (Figure 2.2), the gas abstraction yields are for example only very poorly correlated to size in Swedish landfills and may vary by a factor 4 or 5.

One is therefore justified in considering whether the reliability of national methane inventories obtained by this "top-down" approach could not be greatly improved by applying selectively certain "bottom-up" results at a larger scale. Useful national or global data sets and data bases should be identified and analysed, so as to suggest corrective strategies and develop reasonable ranges, for each variable of the model and for landfills of various types. For this it is necessary to improve our presently crude estimates of LFG generation, but also to better understand and control the processes which determine the efficiency of gas abstraction, a field were particularly few systematic studies have been carried out and most research and optimisation is carried informally by the landfill operators themselves, which therefore limits the diffusion and exchange of information and improvements.

4.3.2 Carbon dioxide

Though carbon dioxide is present in landfill gas in concentrations analogous to methane, and ultimately a large fraction of the methane generated is actually converted to CO_2 before it reaches the atmosphere, either biologically in the cover soil or by combustion, the emissions of CO_2 from landfills are generally not considered. They are

not relevant in the global CO_2 budgets, when compared to the emissions originating from the combustion of fossil fuels.

However a better knowledge of processes governing CO_2 emissions and their more precise quantification could be useful on a world-wide scale, in the frame of a global assessment of the role of landfills as carbon sinks.

In view of the probable shift towards proportionally higher CO_2 emissions from landfills, CO_2 emissions should also be considered as to their impacts on a more local level on the landfill cover soil and vegetation, as well as the possible interactions of CO_2 in the mitigation processes of other gaseous compounds.

4.3.3 Non methane organic compounds

As shown in Figure 4.3, the NMOC's present in landfill gas are released and emitted through processes which differ notably from those governing the emissions of the major components.

The present trend towards less biologically active landfills suggests less degradation of NMOC's will occur inside landfills, since a recent study has shown that stable methanogenic conditions are favourable to such processes (Ejlertsson, *et al.*, 1999). Decreasing methane production may also adversely affect the degradation of NMOC's by the methane oxidizing consortia, who are be able to co-metabolize a large range of chlorinated and aromatic hydrocarbons. However, as they obtain no energy from these compounds, degradation only occurs in the presence of methane, as has been demonstrated for example in some recent preliminary investigations on landfill cover soil (Kjeldsen, 1996).

Caution must be observed in using methane generation and emission models to predict the emissions of NMOC's, as is for example the case in the US for calculation of NMOC emissions from landfills (EPA, 1995). Preliminary field measurements have shown that this approach could well result in an overestimation of the total NMOC emissions by 2 to 4 orders of magnitude (Bogner *et al*., 1997a).

CH$_4$ and CO$_2$ processes

NMOC processes

Net CH$_4$ emissions or CH$_4$ uptake from atmosphere

Net NMOC emissions or NMOC uptake from atmosphere

Net CO$_2$ emissions

Atmosphere

Additional CO$_2$ from soil respiration and direct oxidation of soil organic C

Soil cover

Adsorption and biodegradation

Oxidation of CH$_4$ to CO$_2$

Refuse

CH$_4$ and CO$_2$ generation from anaerobic microbial degradation of organic C

Volatilization, chemical and biological transformations, adsorption

Figure 4.3   Comparison of processes governing NMOC emissions, as opposed to those governing methane and carbon dioxide emissions (from Bogner & Spokas, 1996.)

A conceptual model for the emissions of organic compounds from landfills was proposed by Öman (1997). Three processes intervening in the fate of these compounds were included in the model: sorption, transformation, through microbial or chemical processes, and volatilisation. Comparisons of emissions predicted by this model were made for seven compounds, with concentrations found in landfill leachate in previous studies, but no assessment of gas-phase emissions was made. The cited modelling approach is too simplified, for example the availability of compounds to microbial degradation may be regulated by physical constraints, but similar tools could however be of use also for predicting gas-phase emissions.

4.4 Conclusions

4.4.1 Landfill processes and gas generation

The observation that landfill gas generation takes place over decades in landfills, whereas the same wastes can degrade in a few days in reactors indicates that one or several factors create unfavourable conditions for biological processes in landfills. A

similar indication is given by the fact that gas generation may be described equally well by zero, first or second order reactions. Thus it is not productive to implant models from other areas on the biogenic transformations of landfilled wastes, and these should be further studied in their own right. Landfill cells can serve both as a practical tool for improved process control and as an instrument for improved observation and modelling. This applies to all kinds of waste disposal, not only for the wastes that are already known to form gas. Many more landfill cells must be established and monitored over much longer times before there is real hope for a breakthrough with regard to our capacity of predicting landfill processes.

There are also many other activities beside landfilling, which could have an impact on gaseous emissions, such as composting and the storage of recyclables. There is a general lack of knowledge concerning the impact of such activities, and it this is a serious deficiency, not only from an environmental perspective, but also in view of the occupational health hazards that may result from such emissions.

4.4.2 Gas abstraction

Gas abstraction is a technology with a great potential for improvement. A lot of information already exists, but it needs to be compiled and critically evaluated. There is room for both full-scale investigations and more fundamental studies for issues such as:

- what is the impact of gas abstraction on gas generation? What is myth and what is truth?

- How is gas transported within a landfill?

- How should landfills and landfill gas systems be designed to optimise gas capture?

- How can gas emissions be routinely assessed in a simple fashion?

- Control of gas abstraction: which analytes should be used for optimal control? Typically, only methane and oxygen are monitored at present.

- How can the available data from gas abstraction systems be used in a more efficient way?

The gas abstraction systems of today may work fairly well under some conditions and less so under others. In particular, there is scope for improvement of systems and situations which are very much influenced by weather conditions. The need for development extends from the design of more efficient abstraction equipment, to the control of the extraction systems, including data management.

4.4.3 Emission measurements: present and future needs

Emissions from landfills, where gas movement is often affected by the existence of a subsurface pumped system, are a combination of both convective and diffusive flows. Subsurface vertical concentration measurements consider only diffusive transport, though the soil diffusion coefficient may be corrected for pressure conditions. Micrometeorological methods, on the other hand, rely on above-ground convection and turbulent transfer processes. Chamber techniques do allow to measure global fluxes, but only over a very small surface and short time period. No methods currently exist which satisfactorily address coupled subsurface/surface processes across the soil/atmosphere interface and future research efforts should be applied in that direction.

Investigations should also be aimed at better understanding how surface processes such as wind, other turbulent forces and barometric pressure changes act in conjunction with subsurface processes, such as infiltration to induce small gas pressure gradients in subsurface soil pores. These are potentially very important driving forces of emissions (Bogner & Scott, 1997).

Comparisons of emission measurements by two or more techniques at a given landfill site, an essential step to validate the existing data sets, are at present very few and concern exclusively methane emissions (Galle *et al*., 1998; Savanne *et al*. 1998). There is a need to obtain more comparative data sets, covering also other types of emissions, such as CO_2, N_2O, H_2S and NMOC's. The combined use of tracers and FTIR seems to hold promise for the site-scale evaluation of the emissions of some individual gases, but probably not for NMOC's. It and other approaches, still to be defined, could be used to create a database of emission ranges for various landfill types and gases, possibly in the form of a global survey of the emissions from Swedish landfills, and also for obtaining information on variations at individual sites. In this way it should be possible to significantly improve the present global estimates of emissions from landfills, in particular for methane (see 4.4.5).

The measured methane emissions obtained to date have underlined the wide variability in space and time of such fluxes. In some case, landfills have even been shown to act as net sinks for atmospheric methane. Sampling designs for landfill emissions should incorporate measures of heterogeneity and should be always be tempered with an understanding of the influence of important controlling variables, such as soil type, moisture status, cover design or presence of gas recovery wells. Emission modelling experiments such as that of Czepiel *et al.* (1996), involving whole landfills should conducted on other landfills, varying in size, design and waste composition and be enlarged to other gases and other potential mitigation processes.

4.4.4 Optimising emission control

Studying emissions and mitigation processes at site-specific scale is essential for emission control. There is a need to assess the efficiency of cover materials and designs, both those used as intermediate covers and the final capping of closed landfills, with regard to their mitigation potential for methane and other environmentally significant compounds. Results obtained to-date on the methane oxidising potential of various types of soils are encouraging. Some preliminary investigations have also been conducted on the abatement of other compounds by methane-oxidising consortia and confirm the co-metabolisation capacities of these microorganisms. Because of the complex composition and variability of landfill gas, it is essential that more trials be carried out under real field conditions. There is also the need to address landfill emissions of other trace gases (reduced sulphur compounds, nitrous oxides and halogenated hydrocarbons, especially volatile organic compounds involved in tropospheric ozone formation) and the possible interactions between these and methane. Studies of the net long-term effects of methane oxidation combined with known inhibitory mechanisms at representative field sites need to be initiated.

Future research should address the question of designing optimal top covers for emission control, but should also investigate the potential of using biofilters as substitutes for flares, to treat LFG in cases where energy recovery is not economically feasible. In view of the foreseeable evolution towards less biologically active wastes going to landfill, more and more landfills will be producing smaller amounts of gas of low combustibility, but not necessarily environmentally harmless. Biofilters could prove to be a low-cost and adaptable solution.

There is also a need to better understand how the landfill design and geometry impact on emissions and how this could be used as a part of an emission control system. Passive (*i.e.* landfill design, top cover optimisation) and active (gas abstraction, biofilters) measures for emission control need to be better integrated and their interactions should be further studied.

4.4.5 Improving national estimates of methane emissions from landfills

Unlike other important area sources of atmospheric methane, landfills have not been the subject of a co-ordinated international measurement and modelling program. Such a program is needed to reduce data uncertainties and provide the basis for improved models at various scales. This is especially important for closing major gaps between current "bottom-up" and "top-down" methods and results. There is a need to develop reasonable ranges for emissions at larger scale for landfills of various types. A larger database of landfill site measurements is needed to determine fluxes representative of various cover materials, topography, and climatic conditions. Major resources are needed to measure whole landfill emissions at larger representative sites with various management practices and to address specific problems at smaller sites (especially older

sites without engineered gas control and less-managed sites in developing countries). It would therefore be more efficient if efforts could be co-ordinated at an international level and the already existing informal network and co-operation strengthened.

One possible approach for improving estimates of methane emissions is the use probability density functions to evaluate net methane emissions from a larger database of stratified site measurements. However, given the heterogeneity of municipal solid waste, there is a need to quantitatively model individual categories of wastes in order to track temporal trends. In addition, there is a need to forecast emissions based on different waste management scenarios, which will result in different characteristics of the waste being landfilled.

5. Concluding remarks and recommendations - towards sustainable waste disposal

The number of steps between the generation of waste and its arrival in a landfill is tending to increase. More and more sorting, diversion and processing stages are being introduced, aiming at reducing the amounts and reactivity of the waste before it is disposed. Following this tendency, the amounts of landfill gas that are formed in the landfills will probably decrease over the next decades, but it will not be reduced to zero. On the other hand, there are many other activities that are happening at landfills or elsewhere that may cause gaseous emissions, such as composting and intermediate storage of recyclables. Furthermore, we must now account for new landfill types, receiving waste pre-treated in a variety of ways and for which often little or no data is available. Finally, the present trend towards less biodegradable waste reaching landfills could be overestimated and may also change over the coming years.

The recent legislative trends towards a limitation of degradable organic materials, that may be landfilled in the future (2005), should result in a substantial lowering of the gas formation in landfills over the next 40-50 years. Eventually this would render LFG abstraction unnecessary at most landfills and thus limit the demand for research in this area. However, this legislation has not yet come into force and it is not proven that it will be successful. No thorough evaluation has been found in the literature of whether the final impact of this legislation will be beneficial to the environment, but it is just as unsubstantiated that its impact will be negative.

Contrary to the trend observable in many European countries, there is no tendency to avoid landfilling of organic materials in the USA, but rather, a number of projects are underway to develop landfill treatment systems for organic wastes. This is of interest for several reasons. To begin with, USEPA has supported extensive development work in life-cycle analysis (LCA) of integrated waste management during the last years, and if landfill treatment of organic waste was so detrimental for the environment, one would expect that the evaluations undertaken would have shown it, which they have not. Another reason why the American position is interesting is the fact that the LFG potential of the USA is much greater than that of the EU, in spite of the larger population of the European Union. In a global perspective, since the majority of the LFG-formation potential is gradually shifting from Europe and America to developing economies in Asia, Africa and Latin America, the probable decrease of LFG emissions from European landfills may well be annulled by larger increases elsewhere. It may prove more productive to try to improve LFG management in developing countries, than to impose new restrictions on European landfills.

An increase of waste minimisation and recycling has been taken for granted in the recent years, but it is only feasible with the consent and voluntary participation of the citizens. As in all fields where public opinion is an essential variable, temporal variations are to be expected. There may be a decrease of public acceptance due to the constraints that increased recycling imposes, as well as an increase. Furthermore, past experiences indicate that the composition of landfilled wastes is not affected much by recycling, so the ban on landfilling of organic material is potentially a more decisive change.

In conclusion, one can outline two extreme scenarios for the management of not recycled organic materials in the next twenty years, neither of which will probably actually come entirely true, but which may nevertheless help to understand the situation:

A   The "burn and stabilise" scenario

B   The "treat at landfills" scenario

Scenario A may also be called the Swiss scenario, since it is in this country that both legislation and actual development have already progressed furthest in Europe. To date, Switzerland incinerates 80% of its unsorted municipal waste, which constitutes about 60% of the total MSW, while the remaining 40% are collected separately and recycled (www.admin.ch/buwal/e/themen/umwelt/abfaelle). Landfilling of any wastes that are combustible will be prohibited as of the 1st of January 2000, though this ban is now being contested by some local authorities. Scenario B is perhaps at present most advanced in Sweden and is probably developing most in countries like the USA, the UK and Australia.

Scenario A should result in decreased volumes of LFG generated and in some cases the landfills could in effect serve as a carbon dioxide sink that would in a very minor way compensate for the increased CO_2 emissions generated by incineration. Under this scenario, the relative importance of landfills as sources for atmospheric CO_2 and CH_4 would decrease and other gas and particulate emissions from landfills to the atmosphere would become proportionally more important. There is no evidence that the emissions of other compounds would increase with a reduced biogas production, but there is no evidence to the contrary either. The lack of information merits further investigation. Furthermore, this slow trailing off of gas generation precisely does not signify that LFG would suddenly stop being emitted, but rather that maintaining and developing our knowledge about LFG management may suddenly appear to become less interesting economically. However, there would still be a need to increase our knowledge about the end-phase degradation of landfilled waste and to develop technically sound and economically viable systems for the management of emissions under conditions of declining gas generation.

Under scenario B, there is an obvious need to intensify the development of landfill technology, in order to gain a better control of the potential LFG emissions. Bioreactor landfills or biocells will most likely be a key feature in such a scenario. There are a number of issues regarding the design and operation of biocells which merit research and technical development. This appears obvious if one considers the improved gas yields that recent research programs have shown to be possible, compared the slow degradation rates normally observed in "extensively" managed landfills. It is important that the stabilisation of landfilled waste take place within a few years of its disposal, in order to concentrate emissions over a short time period, thus improving gas recovery yields, reducing emissions and the costs of emission control. Thus, this scenario includes the need of developing of both pre- and post-disposal technology for organic materials that are treated in landfills.

Some R & D on landfill gas emissions will be needed regardless of which of the above scenarios is the closest approximation to future developments. The theme is a better understanding and prediction of landfill processes in general, and its application for different purposes. There is a need for:

- better models of LFG generation and emission;

- better knowledge about emissions other than methane and carbon dioxide;

- better knowledge both about emissions from, and processes occurring in, other types of landfills than the ordinary MSW landfill;

- better knowledge about the working environment of landfills;

- better tools for the environmental impact assessment of various waste management options.

Present models on landfill gas generation and emission rely heavily on assumptions based on highly engineered bioreactors and rapid turnover rates. However, if one is looking to design landfills for long-term sustainability, it is necessary to create new models, specifically developed for landfills, which integrate the different temporal and spatial scales of the processes occurring in the landfills. Such models, using for example the power and versatility of GIS and multivariate analysis, could firstly constitute important aids for improved landfill management and secondly, also be used as better predictive tools.

Sustainability also means a healthy working environment. Gas-phase emissions certainly constitute an important variable affecting the occupational health of landfill workers, but as yet, very little research has been carried out in this field. In the same line

of thought, to be sustainable a waste treatment plant should not become a source of nuisance to its neighbours. The odour potential of the new handling procedures which are expanding, as well as of the residues of these procedures destined for disposal, should be carefully assessed, and, if necessary, adequate avoidance or abatement measures developed.

The available and to-be-developed understanding of landfill processes and waste characteristics should be used to improve evaluation tools such as LCA. Comparative studies of landfills versus other treatment systems and assessments of the global impacts of different waste management scenarios should also be undertaken.

Since LFG emissions are a global problem, we must also direct our attention to the world situation, and see what can be done to help improve landfill technology in developing countries.

This issue can be addressed in numerous ways, and the Swedish Environmental Protection Agency could for example contribute towards improving the present level of information by supporting research and development projects which include international collaborations, by assisting in the creation of information networks, and improving the availability of data (statistics).

Networking is essential since the world-wide landfill gas R & D community is fairly small. Many mistakes (and costs) could be avoided if communication was improved, this could be enacted through travel grants, www-conferences and workshops.

If more process relevant data was demanded and presented, the usefulness of the statistics concerning landfills could be improved, so that such data could be exploited, not only for inventorying wastes and emissions, but also towards acquiring a better understanding of the actual functioning of the landfills.

Research should be both academic and applied, but it is important to base investments in new knowledge on the actual needs of the waste management sector, so a close link to the waste management business is essential. The funding of "industrial PhD student" positions in the waste management companies could be one way of ensuring a good interface between the research and management sectors.

In conclusion it should not be forgotten that the knowledge won through research is futile and may be lost if it is not distributed and applied. There is already much knowledge available, both from research and practice, that has a bearing on the gas

emissions from landfills. It is important that the distribution and implementation of this knowledge be supported in various ways.

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