METHANE EMISSIONS FROM LANDFILLS: OPTIONS FOR MEASUREMENT AND CONTROL

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SUMMARY : Organic matter in waste deposited in landfills creates an anaerobic environment, generating landfill gas (LFG). Among the potential hazards with LFG are: 1) Fires and explosions; 2) Odours and toxicity; 3) Damage to vegetation; 4) Climate effects (greenhouse and smog); 5) Water pollution. LFG normally consists of >50% methane, which is a serious greenhouse gas, but also an energy source. Due to the LFG methane burden on the atmosphere, there is a need to know the methane emissions from landfills. International and regional methane emissions have hitherto been calculated from potential LFG production using data from laboratory-scale incubations of different waste fractions. It is essential to use data from field measurements. Such methods, measuring total methane emissions from landfills with good precision, have now been developed. There are also methods available for estimation of methane oxidation, through direct measurements of carbon isotopes.

1. INTRODUCTION

Methods of managing household and industrial waste differ substantially among different countries. Landfilling is still the predominant method, especially in developing countries, but even in Australia, the United Kingdom, and the Mediterranean countries more than 85% of municipal solid waste is landfilled (IPCC 1996).

Landfilling of waste is inescapable today. Some fractions can neither be recycled nor burned. Ashes from incineration of waste are usually buried, similarly to ashes from coal burning. Ashes contain minerals including heavy metals, but also considerable amounts of organic matter (1-2 % organic carbon; Pavasars 1999), which might make the ashes difficult to stabilize using conventional landfill techniques (Bendz et al 1999).

Among environmental problems with landfills, the greenhouse effect is likely to be the most serious due to the CH_4 content of the landfill gas (LFG), which contribute to the increase in the CH_4 concentration of the atmosphere. Beside the contribution to climate change, CH_4 also brings about risks for fires and explosions. Other components in landfill gas can cause odour problems (Young and Parker 1983). The easiest measure to prevent these negative effects of LFG is the installation of active gas recovery systems. Today 30% of the CH_4 produced in Europe is exploited (Meijer 1999 pers. comm.). For 1997, it is estimated that 1.8 Tg methane was utilized for energy purposes in the USA, while 11.6 Tg was emitted (USEPA 1999). However, the number of landfills with gas recovery installations have been steadily increasing during recent years (Thorneloe et al 1999), and the amount of methane recovered in the USA is likely to be higher today.

In view of the trade with quotients (rights to pollute), currently occurring between countries, it is important that estimates of methane emissions are made in a correct way. This is also true for the possibilities to evaluate the potential for energy utilization. In the following, existing methods of determining methane emissions from landfills are discussed. Moreover, we address the biological production and oxidation of methane in landfills, and we also discuss extrapolation of field data for regional and global budget estimates, as well as possible measures to reduce methane leakage from landfills.

Due to the complexity of the factors affecting the main three processes regulating the LFG emissions, modelling is necessary for the understanding of 1) the emissions (where LFG is transported either vertically through the cover or through so-called lateral migration); 2) the gas recovery through extraction; 3) the methane oxidation. These three processes form the basis for extrapolation models leading to estimates of national, regional and global emissions.
Today, the lack of reliable data is an important constraint for such efforts. Thus, ways to circumvent this problem are being investigated. As an example, Bogner and Matthews (1999) presented a model, in which the global contribution of methane from landfills was extrapolated from energy consumption per capita, which was proportional to the amounts of generated waste. Their calculations were obstructed by a lack of data on gas recovery, for which actual statistics are missing at present.

2. METHANE PRODUCTION IN SOLID WASTE

Soon after waste materials have been deposited in landfills, they are subjected to anoxic conditions. This is the result of low oxygen diffusion into the waste in relation to its consumption by micro-organisms utilising the organic matter of the waste as an energy and carbon source. The rates of these processes will depend on the amount of organic matter, the composition of the waste material and the extent to which the newly deposited material temporarily is covered and/or compacted. Thus, easily degradable organic matter leads to a faster microbial growth and oxygen consumption than more resistant materials. Covers and compaction lead to lower penetration rates for gases into the waste and thus limit the availability of oxygen. The anoxic conditions result in the development of an anaerobic fermentative population of micro-organisms, which in close interaction hydrolyse organic polymers and ferment the hydrolysed matter to products which form the substrates for the organisms responsible for methane formation. Without compaction or covers, more oxygen is available, which leads to an extended oxic degradation of the waste. This is what happens in the so-called "open dumps", which is the case for half of the World's landfills (Moore et al 1998). Such landfills run a high risk of spontaneous combustion due to heat development during the aerobic degradation. Normally, a steady rate of methane production is reached after 80-500 days and is then maintained for 10-20 years (Moore et al 1998). The time required for degradation of waste in landfills and the amount of gas formed depends on a number of factors, such as type and amounts of waste, water content, compaction, leachate treatment etc. (Farquhar and Rovers 1973, Rees 1980). The composition of waste differs considerably between different parts of the world. While the waste in developing countries is dominated by garden material, the waste in industrialized countries is dominated by paper (Moore et al 1998). Paper has a higher content of degradable organic carbon, corresponding to a higher methane generation potential. The water content is of the utmost importance and the effect can be both suppressive (Grischek et al 1999) and promotive of methane production in dry waste (Vroon et al 1998).

To arrive at a correct estimate of methane production in a landfill, it is necessary to know the portion of the waste which can give rise to methane (the potential), and the rates of waste degradation and methane production. Steyer et al (1999) investigated several parameters to use for the modelling of waste degradation rates. They concluded that the cellulose and water contents in the waste, as well as the settling of the waste, could be used to estimate the velocity of degradation and thus the gas production. Below we discuss the models generally used for the extrapolation of gas production to national and global levels.

2.1. Models for prediction of LFG production

2.1.1 Static models: IPCC and others

Early estimates of methane emissions from landfills to the atmosphere were built solely on results from a few laboratory experiments of degradation of different waste fractions. Thus, in the calculation by Bingemer and Crutzen (1987), it was assumed that the ratio of organic matter in landfilled waste corresponded to a certain amount of methane. The models of today are also developed from this approach. The IPCC model for estimation of national methane budgets for landfills (IPCC 1996) has become the common standard (e.g. Frøiland Jensen et al. 1999).

The model is fairly simple to use, but is "static": The waste landfilled one year is assumed to be converted to its full potential of LFG and the methane produced is assumed to be released the same year. Thus, if the amount of landfilled waste is increasing, it will have an immediate effect in the model, despite the fact that it may take some years before the methane production reaches its maximum. This approach may be desirable from a political point of view, since it gives a strong incentive for the installation of gas recovery systems. In order to adjust the model towards more real conditions, several conversion factors (for example 1.0 for managed sites, 0.8 for unmanaged deep and 0.4 for unmanaged shallow sites etc.) have been added to later versions of the IPCC model. However, these factors have tended to become more and more difficult to motivate and use.

2.1.2 First order kinetics

For the national LFG budget for Great Britain, a decline of methane production according to first order kinetics was assumed (Aitchison et al 1996). Similarly, some kind of decline mode has been introduced in other national models, e.g. Denmark, Norway, the Netherlands and the USA (Frøiland Jensen 1999). In Sweden, both an earlier budget made by the Swedish Environmental Protection Agency (SNV; Montelius 1997) and in the on-going work at Statistics Sweden (SCB; Rolf Adolfsson, pers. comm. 2000.02.06) consideration has been given to a decline. It is assumed that the gas production is proportional to degradation of organic matter along first order kinetics as described by Gendebien et al (1992; p. 352):

$$C_t = C_0 e^{-kt} \qquad \text{(Eqn. 1)}$$

where C_t = the concentration of organic matter at time t, C_0 = the initial concentration of organic matter, and k = a constant, indicating the half-life (=ln0.5/-k).

An evaluation of different models for methane production in a number of Dutch landfills was carried out by Oonk and Boom (1995). They found that a first order model was the most useful. The k-factor was estimated at 0.094 yr^{-1}, i.e. a half-life of a good 7 years. Kruempelbeck and Ehrig (1999) published preliminary results from investigations of 50 landfills in Germany, where the half-life was estimated at approx. 4 years. Aitchison et al (1996) used k=0.05 yr^{-1} for calculations of the methane production in U.K. waste, which would give a half-life of almost 14 years.

The three examples given above, of different results from the same type of degradation modelling, illustrate either that the conditions for LFG production are entirely different among countries, and/or that the assumptions are too crude. The latter case is supported by observations by Lagerkvist et al (1997). They reported on the methane production in twelve test cells in three different Swedish landfills: After five years none of them had shown any decline, but rather a more or less stable production during the experimental period. Some of the test cells in Brogborough, U. K., also showed a continuous increase in the methane production, and even after 8 years there was still no decrease in the gas production of the six cells investigated (Caine et al 1999). Obviously, each landfill site has to be individually assessed to apply the most suitable model.

3. METHANE EMISSIONS

3.1 Measurement techniques

The traditional field method for measurement of landfill methane emissions is the use of static chambers. They have been placed on the landfill surface with an open part attached to the surface and the accumulated methane concentration in the closed volume has been measured. The method is simple, but laborious if the total emission rate of a landfill is required. This is due mainly to spatial heterogeneity of the landfill cover. However, the method is suitable only for comparisons between different parts of a site, or for following dynamic changes as governed by climate and other factors. Most of the methane escapes from a few weak parts of the landfill cover, which are mostly difficult to identify and measure. Furthermore, recent investigations have indicated that these "hot spots" move over time (Börjesson et al. 2000a). The reasons may be that the intensity of the methane production moves between different parts of a site, depending on the composition of the waste and for how long the degradation has proceeded and on changes in the landfill cover material due to moisture differences. This affects the gas diffusion characteristics and thus gas transport and probably also methane oxidation by bacteria in the surface soil.

Table 1. Estimates of total methane emissions from landfills with household waste.

Site	Area (ha)	Amount of waste (kg)	Emissions ($g\ CH_4\ m^{-2}\ h^{-1}$)	Reference
Lübars, Germany	29	?	1.9 [a,b]	Jager and Peters 1985
Moscow, Russia	60	($24 \cdot 10^6\ m^3$)	0.60 [a,b]	Nozhevnikova et al 1993
Tokyo, Japan	200	$31 \cdot 10^9$	8.3	Tohjima and Wakita 1993
(unknown), France	3	?	0.44	Pokryszka et al 1995
Oak Ridge, USA	7	?	0.27	Hovde et al 1995
Nashua, USA	24	$2 \cdot 10^9$	2.58, 2.80 [b,c]	Mosher et al 1996
18 sites, The Netherlands	1.7-30	$0.17\text{-}2.3 \cdot 10^9$	0.05-10.2 [b]	Oonk and Boom 1995
Hagby, Sweden	0.4	$10 \cdot 10^6$	0.90	Börjesson and Svensson 1997
Nauerna, The Netherlands	60	$5.4 \cdot 10^9$	0.375	Scharff and Hensen 1999
Randy-Condé, France	8	?	0.06-3.7 [d]	Trégourès et al 1999
Falköping, Sweden	3	($0.3 \cdot 10^6\ m^3$)	1.25	Galle et al 2000

[a] Recalculated from annual values

[b] Conversions were made assuming that 1 m^3 CH_4=1 Nm^3 CH_4=0.656 kg CH_4

[c] Two methods were used: chambers/geostatistics and tracer gas

[d] Seven different methods, comprising three with chambers and four remote sensing techniques

The existing data on whole-landfill estimates of methane emissions in the open literature are compiled in Table 1. The first report by Jager and Peters (1985) was based on measurements with static chambers placed on what were considered to be representative areas during different times of a season. Börjesson and Svensson (1997) used chambers placed in a transect. Static chambers were also used by Nozhevnikova et al (1993) and Mosher et al (1996), with the chambers arranged in a grid pattern in order to enable geostatistical treatment (kriging) of data for integration of the individual chambers. This type of statistics was also used by Pokryszka et al (1995), who employed a dynamic chamber (with a inert sweep-gas flowing through). Examples of remote sensing and over-ground techniques are a moveable flame ionisation detector (FID) (Tohjima and Wakita 1993), and diode lasers (Hovde et al 1995; Scharff and Hensen 1999). Tracer gas has been used in combination with FID (Mosher et al 1996; Trégourès et al 1999) and in combination with FTIR (Fourier transform infra-red)-analysis (Galle et al 2000). Micrometeorology in combination with FID was used on 18 sites in the Netherlands by Oonk and Boom (1995). Their report is the first in which a national budget has been calculated from field data. They estimated Dutch methane emissions from landfills in 1993 at 282 Gg (364 formed, 51 recovered, 31 oxidised) within a range of uncertainty of 170-405 Gg. This lead to a reduction of an earlier budget by 25%.

3.2. Comparative studies of measurement techniques

Czepiel et al (1996) have made comparative studies, and have also published a budget for the North-American state New Hampshire. This group have later published a similar study (Mosher et al 1999). In both these reports it was concluded that static chambers (in combination with geostatistics) and a remote sensing technique with tracer gas/FID gave comparable results. Both Oonk and Boom (1995) and Trégourès et al (1999) reported that micrometeorological methods gave lower values than chambers. Trégourès et al (1999) compared seven different methods of measuring methane emissions. Two types of laser diodes gave lower values than the other methods. Chambers, FID and FTIR gave similar results. Galle et al (2000) used tracer gas in combination with optical measurement (FTIR), which showed approx. 4 times higher emissions than a comparative estimate based on chambers/geostatistics (Börjesson et al 2000a). The difference was probably due to a too crude grid for the chambers in order to cover the "hot-spots", i.e. locations with the highest fluxes. Drawbacks with chambers are also that the results contain an enormous variation. Apart from the limited reliability, another important aspect is the time-consumption. In this respect, the chambers are also inferior to the remote sensing and above-ground techniques (Oonk and Boom 1995, Czepiel et al 1996, Börjesson et al 2000). Remote sensing techniques are obviously outstanding and today they are the only reliable methods of determining total emissions of methane from landfills.

4. METHANE OXIDATION

4.1. Methane-oxidising bacteria

Parts of the methane diffusing into landfill cover soils may be oxidised by methanotrophic bacteria which use the following reactions to gain energy and carbon for their growth (Hanson and Hanson (1996):

$$CH_4 \rightarrow CH_3OH \rightarrow HCHO \rightarrow HCOOH \rightarrow CO_2 \qquad \text{(Eqn. 2)}$$

Energy is yielded in all steps, except in the first. The important intermediate formaldehyde (HCHO) can be used by the bacteria for synthesis of new cell material. HCHO can also be transformed and stored as polymers. Polymers can also be excreted, sometimes in amounts so

Table 2. Estimates of methane oxidation in landfills made with ^{13}C-analysis.

Site	Number of landfills	Method	Ratio of CH_4 oxidised (% of emissions)	Reference
Germany, The Netherlands	2	chambers	39 / 46%	Bergamaschi et al 1998
New Hampshire, USA	6	plume	10%	Liptay et al 1998
New Hampshire, USA	1	plume	12 ± 8%	Chanton et al 1999
Florida, USA	1	chambers	20 ± 3%	Chanton and Liptay 2000
Sweden	2	chambers	>20%	Börjesson et al 2000b

huge that the methane-oxidising bacteria themselves are hampered (Hilger et al 1999).

Through the use of PLFA (phospholipid fatty acid)-analysis it has been shown that methane oxidation in landfill covers could be linked to the two main types of methanotrophic bacteria, but not in an easy-interpreted pattern (Börjesson et al 1998a). Molecular biology methods have recently been developed, which enable a determination of methane-oxidising organisms in soil samples (Wise et al 1999), but quantitative measures have not yet been made with these techniques.

4.2. The significance of methane oxidation in landfills

Comparisons of the methane-oxidising capacity, measured in incubations of soil samples with methane in excess (Whalen et al 1990, Börjesson 1997) and in column experiments (Kightley et al 1995), have given similar results, with an oxidation capacity between 0.14 and 16.8 gCH_4 m^{-2} h^{-1}. These capacities would be enough to take care most of the methane that is produced inside the landfills, but obviously this is not the case. Recently, ^{13}C-techniques for the estimation of methane oxidation have been developed for utilisation in landfill studies. This is probably the best methodology at present. The method (as described by Liptay et al 1998) draws upon the fact that methanotrophs prefer methane containing the common, light isotope ^{12}C and discriminate against methane containing the heavier ^{13}C. Thus, methane oxidation can be estimated through comparison of the content of ^{13}C in methane emitted from the landfill surface with ^{13}C in methane found inside the landfill (in the anaerobic part). Knowledge is required about how large this discrimination is, i.e. the fractionation factor α_{ox} must be determined. This factor varies with soil type and temperatures. Similarly to measurements of total emissions with tracer gas techniques, ^{13}C-analysis should also be done from methane in the plume, because it is important to include the "hot spot" fluxes. Otherwise there is a risk for an over-estimation of the oxidation. A comparison of values obtained from chamber and plume measurements indicates a tendency for plume values to be lower than values from chamber measurements (Table 2).

Among climatic factors, temperature is probably the most important regulator. Experiments with ^{13}C have shown that no methane oxidation occurs at atmospheric temperatures below 0°C (Chanton et al 1999, Börjesson et al 2000b). From these results it was also obvious that oxidation occurred only in the surface layer.

5. POSSIBLE MANAGEMENT PRACTICES TO REDUCE GASEOUS EMISSIONS FROM WASTE

A very high efficiency of preventative measures is required to justify landfilling as a management practice for organic wastes: If methane is calculated as CO_2-equivalents (one methane molecule corresponds to a global warming effect by 21 carbon dioxide molecules in a

100-year perspective), and we assume that half of the landfilled carbon will give rise to methane, more than 80% of the methane produced must be taken care of, i.e. recovered and burned or oxidised.

Options:

i. Direction of the organic waste from landfills towards incineration, composting or anaerobic digestion of the organic matter will lower the CH_4 burden on the atmosphere.

ii. Gas recovery is the most important measure to reduce methane emissions from already landfilled waste (40-90% efficiency; Augenstein and Pacey 1991, Oonk and Boom 1995, Börjesson and Svensson 1997, Börjesson et al. 2000a). Such an efficiency can never be replaced by a methane-oxidising cover material (10-46% efficiency; cf. Table 2). Combustion in kilns is preferable. Flaring is less acceptable, due to risks with low efficiency causing dioxin formation. Gas recovery can also be made more efficient than is the case at many landfill sites today. An efficiency of 50% is not acceptable. An effective drainage system should be installed as early as during the initial phase. Commonly, drilling and installation of vertical pipes is carried out after the landfill is completed. This means that the methane production is established and that lots of methane has already escaped, but also that the drainage will be much less effective because drainage pipes are often placed at wide intervals.

iii. Covering. Since the methane-oxidising bacteria are not active at temperatures around or below 0°C, methane will flow unaffected through the cover at low temperatures. Thus, during winter the type of cover material does not matter. Compost is often suggested for landfill covering, because it has a high methane oxidation capacity (Figueroa 1993, Börjesson et al 1998a),. However, compost of good quality is expensive to produce, and has alternative areas of use such as fertilisation, garden soils etc., which can be quite profitable for the landfill-owner. The compost materials used for landfill covering in Sweden today are often contaminated with heavy metals, petroleum residues etc., which inhibits methane oxidation (Börjesson unpubl.). These kind of soils should preferably be buried. A porous material would allow more oxygen to diffuse, which would promote methane-oxidising bacteria, but it would also let through more methane (Börjesson et al 1998a). The only reason to use compost would be to prevent odour. It is known that organisms in compost can effectively take care of malodorous compounds such as mercaptans, carbon disulphide etc. (Muntoni and Cossu 1997). Apart from the odour problems, which could be solved through an initial covering with organic matter, the hydrological aspects should be guiding for the choice of cover materials. Both from the water- and gas leakage points of view, materials prone to developing cracks and fissures should be avoided.

iv. Landfill mining. Landfilled waste is dug out. After the recycling of valuable fractions (e.g. metals), the hazardous waste is eliminated through incineration (Kornberg et al 1993).

6. CONCLUSIONS

6.1 Options for Supervision

• Gas recovery projects should be supported, and existing extraction systems could be made more effective.

• Emissions are best measured with remote-sensing techniques - where the geography and costs allows for it. Cracks and fissures can also be detected with "sniffers", e.g. Gas-Trac® and similar instruments. Additional covering should be done where thin layers are observed.

• Methane-oxidising capacity in soils and similar cover materials should be tested. Additional covering could be required. A 1 g dry weight soil sample from a one-year old cover should oxidise at least 10 µg CH_4 per hour.

6.2 The Need for more Research

Many questions remain to be answered:

1. Methane production

More data is needed on degradation rates for different types of waste under realistic conditions. Landfills with different climatological and geological conditions should be investigated. Above all, data from warmer regions are lacking completely. As mentioned, global data from gas recovery are also missing. This information is needed in order to enable modelling and up-scaling. The field data produced to date on methane emissions from landfills are also difficult to interpret due to the lack of uniform variables. An example of this is given in Table 1, which shows that the methane emissions measured in various investigations have been reported, but the methane-generating amounts of waste on the actual landfill sites are seldom known.

2. Effectiveness of different types (strategies) for gas recovery

Vertical and horizontal drainage systems, distance between pipes and wells, pump pressure etc. need to be investigated further. It must also be established whether existing landfills can be much improved at a reasonable cost.

3. How important is methane oxidation?

We know very little about the significance of methane oxidation in warmer climates, so data are required from these areas. In addition, more information is needed about the establishment of methane-oxidising organisms in different types of cover materials.

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This paper was first presented at the Waste 2000 Conference, Stratford-upon-Avon 2-4 October 2000 (Proc. pp. 31-40), and is reused by kind permission.