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> > TELLUS

# A national landfill methane budget for Sweden based on field measurements, and an evaluation of IPCC models

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#### ABSTRACT

Seven Swedish landfills were investigated from 2001 to 2003. On each landfill, a measure of the total methane production was calculated from data on: (1) methane emissions (leakage); (2) methane oxidation and (3) from gas recovery. Methane emissions were determined via a tracer gas (N<sub>2</sub>O) release-based remote sensing method. N<sub>2</sub>O and CH<sub>4</sub> were measured with an Fourier Transform infrared detector at a distance of more than 1 km downwind from the landfills. Methane oxidation in the landfill covers was measured with the stable carbon isotope method. The efficiency in gas recovery systems proved to be highly variable, but on an average, 51% of the produced landfill gas was captured.

A first-order decay model, based on four fractions (waste from households and parks, sludges and industrial waste), showed that the use of a degradable organic carbon fraction (DOCf) value of 0.54, in accordance with the default value for DOCf of 0.50 in the latest IPCC model, gave an emission estimate similar to the official national reports.

# 1. Introduction

#### 1.1. Methane from landfills

In many countries in Europe, landfilling of organic matter is now restricted. In Sweden this was regulated in 2005 (RVF, 2006). Nevertheless, many landfill sites will continue to produce biogas for many years, with a composition of about 50% methane. A portion of these gases will escape capture and contribute to atmospheric greenhouse gas loading, where the global warming potential of methane corresponds to 25 carbon dioxide equivalents (100 yr GWP; cf. Solomon et al., 2007). Estimates of landfill gas emissions are required for the national greenhouse gas inventories made according to the Intergovernmental Panel on Climate Change (IPCC) guidelines (the latest version being IPCC, 2006) and for trading of with greenhouse gas credits, as well as for the utilization of landfill gas emission have been based for the most part on data on the amount of waste deposited.

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Such estimates have been calculated from field measurements to a very limited extent previously (e.g. Oonk and Boom, 1995).

#### 1.2. Field measurements

It is widely recognised that landfill gas that is not recovered through gas extraction generally escapes from a few weak spots in the landfill cover or from leaking pipe systems (e.g. Nozhevnikova et al., 1993; Lewis et al., 2003). Such areas are difficult to identify and measure emissions from. Recent investigations have also indicated that such 'hotspots' move over time (Börjesson et al., 2000). Based on earlier comparisons (Börjesson et al., 2000; Galle et al., 2001), it was decided to use the tracer gas technique for measurements of methane emissions in the present study since this method proved to give more reliable and reproducible results than other methods, for example, the chamber technique. For determination of methane oxidation, it was decided to use the carbon isotope technique, which is the most reliable method for in situ measurements, and can also be applied to plume measurements as described by Chanton et al. (1999).

Hence, including data on gas recovery supplied by the landfill operators, we obtained data on total methane production P for

each individual landfill as

$$P = E + R + M,\tag{1}$$

where

E = emissions of methane escaping from the landfill surface  $(\text{kg h}^{-1})$ 

 $R = \text{gas recovery (kg h^{-1})}$ M = methane oxidation (kg h<sup>-1</sup>)

Assuming that the total methane production P is constant over the year for each landfill and by using the official data on landfilled amounts, the aim was to extrapolate the sum of measured Ps for a number of different landfills to construct an annual national methane budget for Sweden.

### 1.3. The IPCC models

By extrapolating our data, we should also be able to compare a methane budget based on field measurements with models based on statistics on waste amounts deposited in landfills. The most important of these models is the IPCC-model, which is widely used. National reports to the IPCC on greenhouse gases from a variety of sources are made every year. For landfill methane production and emissions, either a default model or a first-order decay (FOD) model were suggested as basis for this work (IPCC, 2001). Since our measurements were done 2001-2003, we decided to use 2002 as the basis. Data on landfilled amounts of waste were collected according to what was required for the IPCC (2001) model, and most of the comparisons will therefore be made between our field data and this model or its Swedish version (Swedish Environmental Protection Agency, 2001; with details in Zuber et al., 2001, cf. Table 1).

The original FOD model is simply a multiplication of different factors. The total methane production during a certain year T from landfilled amounts of n waste fractions can be calculated as

Methane production<sub>T</sub> = 
$$16/12 \times F \times DOCf \times MCF$$

$$\times \sum_{t=t_0}^{1} \sum_{X=1}^{n} MSW_{Xt} \times DOC_{Xt} \times t_{tT} \quad (2)$$

The individual parameters are explained in Table 1. In addition to this, a factor for methane oxidation (OX) was also included in the national model (Zuber et al., 2001) and was estimated to be 10% of the amounts that were not recovered through gas extraction. Data on municipal solid waste (MSW) was obtained from officially available statistics, and the factors used as model parameters for degradable carbon are summarized in Table 2.

The factor  $t_{tT}$  in eq. (2) is based on the assumption that the gas production is proportional to degradation of organic matter following first-order kinetics, as described by Gendebien et al. (1992; p. 352):

$$C_t = C_0 \mathrm{e}^{-kt},\tag{3}$$

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 (=  $\ln 0.5/(-k)$ ). 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, including a k-factor estimated at 0.094 yr<sup>-1</sup>, that is, a half-life of 7 yr. Kruempelbeck and Ehrig (1999) published preliminary results from investigations of 50 landfills in Germany, where the half-life was estimated at approximately 4 yr. Aitchison et al. (1996) used  $k = 0.05 \text{ yr}^{-1}$ for calculations of the methane production in U.K. waste, which would give a half-life of almost 14 yr.

The aim of this study was to compare the official model for the Swedish landfill methane estimate with field data, based on the best available methods. For this purpose, we wanted to investigate landfills of different size, age, cover type and waste management practice.

Parameter	Explanation	Value in national estimate (Zuber et al., 2001)	Likely interval for parameter
16/12	Conversion factor for carbon to methane		
F	Ratio of methane in landfill gas, mol%	50%	45%-60%
DOCf	Fraction of degradable carbon dissimilated	0.7	0.5-0.77
MCF	Correction factor for landfill management practice. This has been set to 1, if any activity such as compacting or covering was made.	1	0.95–1
MSW <sub>Xt</sub>	Landfilled quantity of waste type X in the year t (ktons).	According to statistics; see text	
DOC <sub>Xt</sub>	Content of organic carbon in waste of type X in the year t.	Varying (see Table 3)	
$t_{tT}$	$\begin{cases} e^{-0.5\ln 2/t_{1/2}(t-0.5)}(1-e^{-0.5\ln 2/t_{1/2}}), t=1, 2, \dots, T-1\\ 1-e^{-0.5\ln 2/t_{1/2}}, t=T \end{cases}$	$t = 7.5  ext{ yr}$	4–14

Table 1. Parameters for landfill methane production in the IPCC model (IPCC, 2001) and their values in the Swedish version (Zuber et al., 2001)

Site	Coordinates	Waste amounts, ra	nges for 1994–	Household waste, annual average (ktons)	Sludges, annual average (ktons)		
		Household waste	Park	Sludges	Industrial waste		
Filborna (Helsingborg)	56°04N, 12°46E	57.1-69.0	Not reported	1.8-3.8	42.6-82.0	63.31 <sup>c</sup>	2.78 <sup>c</sup>
Heljestorp (Vänersborg)	59°32N, 17°38E	10.5-40.0	0-3.8	1.0-6.7	6.5-42.9	28.27	3.89
Kristianstad	56°01N, 14°08E	4.6-40	0-4.0	0.4-2.4	8.0-89.5	16.46	0.92
Högbytorp (Upplands Bro)	59°32N, 17°38E	20.0-56.5	0.5–3	5-21	90-145.6	30.90	13.11
Sundsvall	62°23N, 17°11E	4.0-10.6	0-0.2	1.6-8.0	19.0-48.2	7.55	4.18
Hagby (Vallentuna) <sup>a</sup>	59°28N, 17°58E	20	Not reported	4.6-5.0	58-120	20	4.78
Visby <sup>b</sup>	57°38N, 18°21E	10.4-14.5	0-0.8	0.1–0.6	2.3-9.3	12.56	0.36

Table 2. Landfill sites, coordinates and amounts of different waste cumulativly deposited annually, together with household waste and sludges for the period 1994–2002

aclosed 1995: waste amounts reported 1994-95

<sup>b</sup>closed 1998: waste amounts reported 1994-97

<sup>c</sup>1992–2000.

# 2. Sites and methods

#### 2.1. Landfill sites

About 30 landfill sites were investigated as targets for our measurements. Seven sites were chosen as based on geographical location, size, age and management conditions (Table 2). The chosen sites also represent different types of cover materials, including mineral soils with at least 1m clay (Hagby, Visby, Kristianstad), mixtures of sewage sludge and soil (Högbytorp, Sundsvall) and mixtures of wood chips and sludge (Filborna, Heljestorp). None of the sites had synthetic liners. All sites except two were actively used during the measurements (the site in Kristianstad was closed 2002, but after our measurements).

Table 2 summarizes data on landfilled amounts given by the official statistics compiled by the Swedish Waste Management Association (RVF, 2001–2003). In some cases, we were also given access to more detailed data from the individual landfill owners.

#### 2.2. Methane emissions

The time correlation tracer system described by Galle et al. (2001), and modified by Samuelsson et al. (2001) was used for measuring methane emissions. This includes the release of the tracer gas  $N_2O$  and concentration measurements with an Fourier transformed infrared (FTIR) spectrometer system. The system is built on a gas-moderated aluminium platform, housed in a temperature controlled box inside a small van. A telescope mast mounted on the van has a tubing with an inlet at 10-m height, connected to an inert pump, which sucks air from the plume through the gas cell with one volume changed in the cell per recorded spectrum. The measuring system is automatically regulated by a computer, with evaluation and presentation of data

in real time. A GPS receiver records the geographical position of the concentration values.

The tracer gas  $N_2O$  was released from cylinders through a regulator converting the pressure from 200 bar to atmospheric levels. The amount of  $N_2O$  released was determined both by a flow integrator and by weighing the cylinders. Depending on the size of the actual landfill site, the amount of methane emitted and the pattern of the plume, the number of  $N_2O$  cylinders on each landfill varied between two and five and the amount of released  $N_2O$  varied between 5.0 and 12.5 kg h<sup>-1</sup>.

Emissions of methane,  $E_{CH_4}$ , were determined as

$$E_{\rm CH_4} = F_{\rm N_2O} \cdot [\rm CH_4] / [\rm N_2O] \cdot M(\rm CH_4) / M(\rm N_2O)$$
(4)

where  $F_{N_2O}$  is the known release of tracer gas, [] are concentrations in the plume (ppb above background) and *M* are molecular weights.

An example of a measurement of the concentration variations in the two gases in a sweeping plume for about 1.5 h is given in Fig. 1.

#### 2.3. Gas recovery

Data on gas recovery from the landfill operators were based on different principles and the time resolution of data between the sites was highly variable. At Filborna and Kristianstad, both the methane concentration and the gas flow were measured continuously, with an uncertainty of approximately 5%. Filborna was the only site with an active flare, and flared gas volumes were included as gas recovery. For Blåberget in Sundsvall, the gas extraction was given as a mean day value based on gas flow and the partial pressure of methane, but the variation over 4d was less than 2.4% indicating stability in these data. The same approach was used for Heljestorp and Hagby, supplemented by



*Fig. 1.* Methane emission measurement at the Filborna landfill (4 April 2001). Time course change of methane ( $CH_4$ ) and tracer gas ( $N_2O$ ) concentrations above atmospheric levels in the left-hand panel, and the correlation between these values in the right-hand panel.

a couple of manual readings of gas flow and methane ratios in the beginning and in the end of the measurement day. For Högbytorp and Visby, the gas extraction was calculated from readings of utilised energy and conversion to methane amounts by the conversion factor 1 kg  $CH_4 = 16.56$  kWh (Gendebien et al., 1992). Readings were made 1–5 times per day.

#### 2.4. Methane oxidation

The content of  $\delta$ 13C in methane, that is, the ratio between the carbon isotopes 13C and 12C can be used for quantify methane oxidation in situ. Most enzyme systems discriminate against 13C, which means that  $\delta$ 13C is low in methane formed in the anaerobic zone, but substantially higher after the gas has passed through the aerated cover and been exposed to microbial methane oxidation. The isotope method used for landfills (as described by Liptay et al., 1998) draws upon the fact that  $\delta$ 13C in methane can be measured

(1) in the anaerobic zone (e.g. from the gas extraction system);

(2) in the wind plume; and

(3) in the background.

The difference between  $\delta 13C$  in methane from the *B*- and *C*samples gives an excess value ( $\delta_{\text{excess}}$ ). This value is then compared with  $\delta_A$ , that is, the *A*-samples to estimate the methane oxidation. An additional term which is required is  $\alpha_{\text{ox}}$ , the extent to which 13C in the methane has been discriminated against by the oxidizing bacteria. This fractionation factor,  $\alpha_{\text{ox}}$ , varies depending on soil structure, moisture and temperature (Chanton and Liptay, 2000; Börjesson et al., 2007) and has to be determined for each landfill cover by

(4) soil incubations.

Taken together, the samples A-D give us an equation for the fraction of  $CH_4(f_{ox})$  oxidized during the upward transport through the landfill cover soil (Chanton and Liptay, 2000; Börjesson et al., 2007):

$$f_{\rm ox} = \frac{(\delta_{\rm excess} - \delta_A)}{1000 \times (\alpha_{\rm ox} - \alpha_{\rm trans})},\tag{5}$$

where  $\alpha_{\text{trans}}$  is an isotope fractionation factor associated with transport of CH<sub>4</sub>, assumed to be = 1 for landfill covers (cf. discussion by Börjesson et al., 2007).

2.4.1. Gas samples in situ. For determination of  $\delta_A$ , samples were taken in triplicate from the gas extraction system of the respective landfill site on each sampling occasion. Samples from the wind plume and in the background ( $\delta_{\text{excess}}$ ) were taken in triplicate in 100 mL glass flasks with aid of the FTIR system.

2.4.2. Incubations. For determination of the fractionation factor  $\alpha_{ox}$ , at least four composite soil samples (ca. 1 kg) were taken randomly from different locations covering a depth of 0–30 cm, from the surface soil of each landfill site. These samples were treated separately (sieving 4 mm, determination of moisture content and loss on ignition).

The strategy was to obtain data as a function of temperature for the fractionation factor  $\alpha$  for each landfill site. Every soil sample was divided into four portions, with between 50 and 100 g wet weight soil, were transferred to 1150 mL glass jars (Schott, Mainz, Germany) and placed in climate chambers at +3, +10, +15 and +20°C. The glass jars were gas-tight and equipped with a butyl rubber stopper to enable sampling. Sixty millilitres of methane was added at time zero.

2.4.3. Analyses. A time-series for estimation of  $\alpha_{ox}$  was based on the methane consumption pattern, as determined by gas chromatographic analysis (Börjesson and Svensson, 1997), with samples taken from the incubation flasks in intervals from one hour up to 60 d. On all occasions, 10 mL samples were withdrawn and stored in evacuated serum vials for later analysis of  $\delta$ 13C- methane. For each glass jar, at least four representative samples were chosen for the time-series. Analyses on incubation samples were made at the Department of Forest Ecology at SLU in Umeå, Sweden, whereas all the other (air) samples were made by Department of Oceanography, Florida State University, USA (see Börjesson et al., 2007 for further details).

#### 2.5. Meteorological measurements

Meteorological data were collected both adjacent to the gas inlet at 10 m height on the mast (cf. Section 2.2) and on the landfill site, where data were collected continuously on soil temperature (at 5-cm depth), air temperature, wind speed and wind direction (Börjesson et al., 2007).

#### 2.6. Model approaches

In the IPCC (2001) model, amounts of waste are multiplied with factors for degradable organic carbon (DOC). The model used for the third national inventory (Swedish Environmental Protection Agency, 2001) is to most extent described by Zuber et al. (2001), and the parameters are given in Table 1. Among variables different from IPCC default values, one of the most important was a DOCf of 0.70 instead of 0.77, which was attributed to a lower temperature in Swedish landfills (30°C) compared with average. Methane oxidation was set to 10% of produced methane instead of zero, and a FOD with a half-life time of 7.5 yr was adopted from Oonk and Boom (1995). The DOC values for individual waste fractions are given in Table 3.

Comparisons were also made with direct correlations between measured total methane production and amounts of landfilled waste according to the official Swedish statistics.

#### 3. Results

# 3.1. Field measurements—gas recovery and total methane production

In our series of landfill sites, Filborna by far received the highest amounts of organic materials, and also showed the highest rates of methane production (Table 5). During the 2 yr and seven measurement periods at the site, the estimated methane production rates were within 5%.

A more intensive period of measurements were performed at Filborna, coincident with improvements of the gas extraction systems between 23 November and 6 December 2001. Measurements prior to this activity showed a gas recovery of 820 and 832 kg CH<sub>4</sub> h<sup>-1</sup>, whereas following improvement, the measurements showed 987 and 1006 kg CH<sub>4</sub> h<sup>-1</sup>, respectively. Thus, the amount of methane emitted to the atmosphere relative to total methane production decreased from 30%–33% to 20%–26%. For the measurement campaign, half a year later (July 2002), the rate of gas extraction had dropped to 806 kg CH<sub>4</sub> h<sup>-1</sup>, and emissions were 28% of the total production of methane.

For Högbytorp and Heljestorp, both active landfills, higher methane production was observed during the 2002–03 campaigns relative to the 2001 measurement period (Table 5). Of the two closed landfill sites, Hagby showed similar CH<sub>4</sub> production rates for 2001 and 2002, possibly owing to a much larger portion of building waste, whereas Visby revealed a substantial decrease in methane production indicating a decline of 30% between 2002 and 2003. It should also be noted that the measurements in April 2002 and in November 2003 at Hagby coincided with problems with the gas extraction system, which resulted in elevated emissions to the atmosphere.

Three of the landfills—Kristianstad, Blåberget and Heljestorp—were measured only once or twice, mainly due to

Waste type	IPCC (2001) default values	National model (Zuber et al., 2001)	Mean annual deposition 1990–2002 (kton)	DOC (kton)
Household waste		0.18-0.22	1147	230
Paper and textiles	0.40			
Food waste	0.15			
Wood or straw	0.30			
Garden and park waste	0.17	0.17	60	10
Industrial waste (mainly slaughter house)		0.12	1513	194
Building waste		0.035	694	27
Sewage sludge		0.07	546	38

Table 3. Fraction of degradable organic carbon (DOCf) in different waste types included in the models for landfill methane production in Sweden



Fig. 2. Total methane production measured at each landfill site, in chronological order. Error bars are 95% confidence interval for each estimate.

difficult geographical conditions for measurements at appropriate distances. This is also shown by a lower correlation coefficient ( $R^2$  for E) between CH<sub>4</sub> and the tracer gas N<sub>2</sub>O in Table 5.

A weighted mean for the gas extraction efficiency of 51% (*SD* 14%) for the seven landfill sites in this study was calculated from the data in Table 5.

3.1.1. Uncertainties in field measurements. The uncertainties caused by instrumental and analytical performances, together with tracer gas positioning, resulted in an average uncertainty of  $\pm 18\%$  (95% confidence interval) for the estimate of methane emissions from the landfill sites. The precision of the measurement in the experiments at Filborna in March 2003 and in Visby 2002 showed a variation in the emission estimate of 11% (1 $\sigma$ /mean) and 7.5% (1 $\sigma$ /mean), respectively, over a time period of 2–3d.

The precision for the estimated production was also quite good, down to  $\pm 4.2\%$  for the measurements at Filborna (Fig. 2). The confidence interval for production estimates was spread between (-6.0%,+6.2%) and (-33%, +204%) in terms relative to the production. The large spread on the positive side was explained by a large uncertainty in the methane oxidation. This affects the estimated production, since the uncertainty in the methane oxidation values are amplified when the emissions are high, for example, at Hagby 2002 (Fig. 2).

#### 3.2. Gas recovery

*3.2.1. Uncertainties in gas recovery.* The amount of extracted methane was measured in different ways at the landfill sites. At Filborna, the methane ratio was measured in the gas flux with an IR-instrument, which regularly was calibrated with a standard gas. The pressure and temperature of the gas flux is measured

through differentiating the pressure with a throttle-valve. According to the manufacturer, the methane concentration could be measured with a precision of  $\pm 5\%$ , and the precision for flux in terms of Nm<sup>3</sup>h<sup>-1</sup> could be determined within  $\pm 0.1\%$ . A more modest estimate of this uncertainty could be assumed as  $\pm 3\%$ . The resulting uncertainty in the extracted amount of methane at Filborna would then be  $\pm 4.9\%$  (RSS = square sum of input errors). An alternative technique would be to measure the effective outtake at the heat transmission to the district heating system or from a kiln by measurements of temperature and in- and outgoing heat transmission. After discussions with operators and consultants within this field, we judge that the gas recovery can be estimated at  $\pm 5\%$  for the investigated landfill sites (assuming 95% confidence intervals).

#### 3.3. Methane oxidation

3.3.1. Observed data. Unfortunately a large number of samples were spoiled, including almost all samples from 2001. Therefore, some values have been interpolated from other occasions with similar conditions concerning temperatures or type of land-fill (Table 5 with notes). This can be justified as methane oxidation, with few exceptions constitutes a minor part of the total amount of methane produced in a landfill (cf. Table 5). Most of these values, including the  $\iota$ 13C for methane in the landfill gas have been reported earlier (Börjesson et al., 2007).

The large variation in the  $\alpha$  values indicates that the estimates of methane oxidation in situ must be considered with caution (Table 4). The ratio of oxidised methane was estimated to vary between 6% and 43% of emitted methane (Table 5), although it was an obvious tendency for the cover materials on the closed sites (Visby and Hagby) to have considerable higher methane oxidation rates than the other sites in our study.

*Table 4.* Estimates of the fractionation factor  $\alpha$  and its temperature dependence ( $T = {}^{\circ}C$ ); r = regression coefficient

Site	$\alpha(T)$	r	Variability in $\alpha$ at estimates (mean, $1\sigma$ )
Filborna	1.0204-0.000098·T	0.16	0.0047
Högbytorp	1.0232-0.000220·T	0.22	0.0039
Blåberget	1.0243-0.000353·T	0.42	0.0054
Visby	1.0208-0.000239·T	0.38	0.0043
Hagby	1.0266-0.000304·T	0.30	0.0073
Heljestorp	1.0358-0.000664·T	0.58	0.0056
All	1.0251-0.000313·T	0.34	0.0052

Table 5. Landfill methane measurements 2001-2003

The ratio of oxidised methane at Filborna varied between 6.2% and 22% (Table 5), with the lowest values observed in winter (T =  $3^{\circ}$ C) and the highest in summer (T =  $14^{\circ}$ C).

3.3.2. Uncertainties in the methane oxidation estimates. The methane oxidation analysis is complex, and small changes within small signals are used. The measurements consist of a number of different steps, each with its specific uncertainty, and the overall uncertainty is therefore large. The narrowest confidence interval obtained for methane oxidation in our study was (-41%, +67% in relative terms) for an oxidation value of 25% (absolute value), but the confidence intervals were often at levels as high as (-50%, +200%).

The methane oxidation estimates were most sensitive to the variation in the fractionation factor  $\alpha_{ox}$ , which in our case caused

Landfill site	Date	Soil tempe- rature at 5 cm depth (°C)	$E = CH_4$ emission to atmos-phere (kgh <sup>-1</sup> )	$\mathbf{R}^2$ för $E$	R = Gas recovery (kgh <sup>-1</sup> )	$M = CH_4$ oxidation (%)	$P = \text{Total CH}_4$ production (E/ (1-M/100)+R) $(\text{kgh}^{-1})$	Ratio to atmosphere ( <i>E/P</i> ) (%)	Efficiency of gas recovery system (%)
Filborna	4 Apr. 2001	8.5	308	0.94	852	18 <sup>a</sup>	1229	25	69
(Helsingborg)	1								
	16 Nov. 2001	9.5	386	0.94	832	18	1304	30	64
	23 Nov. 2001	3.0	441	0.82	820	15 <sup>b</sup>	1340	33	61
	6 Dec. 2001	3.1	256	0.97	987	6.2 <sup>c</sup>	1260	20	78
	7 Dec. 2001	3.1	361	0.92	1006	6.2 <sup>c</sup>	1391	26	72
	2 July 2002	13.9	346	0.80	806	22	1250	28	64
	10 Mar. 2003	3.6	403	0.65–0.91	939	6.2 <sup>c</sup>	1369	29	69
Högbytorp (Upplands-Bro)	6 June 2001	15.2	258	0.75	140	25 <sup>d</sup>	486	53	29
	11 Apr. 2002-	7.3	393	0.96	202	6.0	620	63	33
	10 Nov. 2003	4.9	382	0.84	291	7.7	705	54	41
Blåberget (Sundsvall)	9 Mar. 2002	-1.9	33.8*	0.50	58.3	15	98	35	59
Visby	13 June 2001	11.6	28	0.97	48	37 <sup>e</sup>	92	31	52
	4 June 2002	18.7	19.2	0.97	39	37 <sup>e</sup>	69	29	57
	5 June 2002	15.2	18.6	0.95	39	37 <sup>e</sup>	68	29	57
	26 Nov. 2003	5.1	12.8	0.88	32.4	38	53	24	61
Hagby (Täby)	18 Apr. 2001	9.9	49	0.77	155	37 <sup>e</sup>	233	21	67
	22 Apr. 2002	14.2	124	0.98	32	37	229	54	14
	13 Nov. 2003	3.0	141	0.97	65.7	43	312	45	21
Heljestorp (Vänersborg)	29 Mar. 2001	6.7	136	0.75	134	6.2 <sup>c</sup>	279	49	48
	22 May 2002	16.7	191	0.82	262	25	517	37	51
Kristianstad	12 Apr. 2001	5.0	43	0.78	117	38 <sup>f</sup>	187	23	63

\*Uncertainty in emission estimated at  $\pm 38\%$  due to topography and difficult situation for measurement.

<sup>a</sup>Ratio of methane oxidation from Filborna 16 November 2001, T = 8.5 °C assumed comparable to 9.5 °C.

<sup>b</sup>Background data for methane oxidation from Filborna 16 November 2001, other samples analysed.

<sup>c</sup>Ratio of methane oxidation from Filborna 28 November 2001. T = 3.1-6.7 °C assumed comparable to 2.7 °C.

<sup>d</sup>Ratio of methane oxidation from Heljestorp 23 May 2002. T = 15.2 °C assumed comparable to 16.7 °C.

<sup>e</sup>Ratio of methane oxidation from Hagby 24 April 2002. T = 9.9-18.7 °C assumed comparable to 14.2 °C.

<sup>f</sup>Ratio of methane oxidation from Visby 26 November 2003. T = 5.0 °C assumed comparable to 5.1 °C.

variability in methane oxidation of between -40% and +72% in terms relative to the estimated mean value of 21.9%. When all parameters were varied at the same time, the methane oxidation was 21.9% within a 95% confidence interval of 9.5% (39.3% in absolute numbers).

#### 3.4. Comparison with models

3.4.1. Extrapolations. A simple and straightforward approach for calculations of methane production is to directly relate landfilled amounts to methane production. A linear regression analysis was made between measured annual methane production (as the dependent variable) and mean annual landfilled waste amounts during the landfills' active period, assuming constant production over the year (cf. data in Table 2). A linear model with production as a function of household waste alone gave a high degree of correlation ( $r^2 = 0.96$ , n = 7), with Methane production (kg) = 0.16 Landfilled household waste (kg). To get a model more applicable to normal-sized Swedish landfills, Filborna was excluded because of its large size. This resulted in a reduction of the response factor; that is, Methane production (kg) = $0.13 \times$  Landfilled household waste (kg) ( $r^2 = 0.94$ , n = 6). Between 1990 and 2002, the landfilled amounts of household waste in Sweden averaged 1068 kton, according to the official statistics (RVF, 2001–2003). When applying this amount of waste to the latter model, it results in an annual methane production of  $139\pm28$  kton CH<sub>4</sub> yr<sup>-1</sup> (95% confidence interval, from a mean error of 0.13 in the regression parameter 0.13). Taking away gas recovery (34 kton during 2002 according to statistics) from this value, and when 10% oxidation is subtracted from the rest, we will get an estimate of methane emissions from Swedish landfills at 95 $\pm$ 21 kton CH<sub>4</sub> yr<sup>-1</sup>.

3.4.2. The IPCC model. The comparison between the measured data and the calculated DOC content according to the IPCC (2001)-model is presented in Fig. 3. The two points in the right-hand side of the panel of Fig. 3 are Filborna in the upper part and Högbytorp below the regression line. According to data from Filborna the industrial waste was measured to contain 25% DOC. If this value is applied for Filborna, we get a different regression line according to Fig. 3. The slope of the regression line at 0.36 (with a mean error of 0.038), corresponds to DOCf = 0.54 when applying the units described in Section 2.6 and the data in Table 3.

When IPCC's standard method (IPCC, 2001) was applied for mean annual amounts 1990–2002 (Table 3), with a conversion to DOC (500 kton yr<sup>-1</sup>), the total methane production would be 182 $\pm$ 38 kton CH<sub>4</sub> yr<sup>-1</sup>. After subtracting the gas recovery from this value and assuming the rest to be reduced with 10% by methane oxidation, it yields an estimated annual emission at 134 $\pm$ 31 kton CH<sub>4</sub> yr<sup>-1</sup> (95% confidence interval). This means that the IPCC model shows an estimated annual emission, which is 41% higher than the estimate made directly from measured values and landfilled waste amounts. When applying the best-



*Fig. 3.* Correlation between measured methane production at the seven investigated landfill sites and DOC-values for the landfill waste obtained from statistics according to the IPCC (2001) model, as used by Zuber et al. (2001).

fit DOCf factor of 0.54 instead of 0.7, the IPCC model arrives at an annual emission, which is only 9% higher than the fieldbased estimate 95±21 kton  $CH_4$  yr<sup>-1</sup>. This is almost in line with the national estimate (Zuber et al., 2001), which arrived at an estimate of 88 kton  $CH_4$  in emitted from Swedish landfills in 2002.

3.4.3. A multiple regression model. Regressions were made by the use of measured total methane production as a function of the different waste fractions (household waste, sludge, park waste and industrial waste) as possible variables. Verifying the example of correlation in Section 3.4.1, household waste was chosen as the most important variable. However, a model with two parameters, household waste as the first and sludge as the second explained 98% of the variation. The negative intercept (-1803 ton CH<sub>4</sub> yr<sup>-1</sup>) was avoided by excluding Filborna as an outlier. This can be argued for, since the waste management practice at this site, for example, shredding household waste, is obviously enhancing gas production. With this measure, the variation will be higher and the intercept halved (Fig. 4).

The factor for sludge is higher than the factor for household waste, but it should be noted that the average amounts of landfilled sludge were more than five times lower than for household waste. Therefore, this model result should be taken with care. Furthermore, the size of the data set is limited and contains variability. Nonetheless, the results appear to indicate that sludge can make a significant contribution to methane production in landfills.

# 4. Discussion

The methods chosen for the field measurements gave highly reproducible results, especially so the FTIR system. The

Fig. 4. Comparison between measured total methane production in six landfille household waste and sludge as the waste variables chosen by the program (Filborna excluded as being an outlier). The equation for the model was y (measured total methane production, ton yr<sup>-1</sup>) =

 $131.2 \times$  household waste (kton yr<sup>-1</sup>) +  $155.1 \times$  sludge (kton yr<sup>-1</sup>) –

903.2 (kton yr<sup>-1</sup>) with  $r^2 = 0.992$  (If the intercept is locked in origo,

the equation will be  $y = 85.1 \times \text{household waste} + 181.1 \times \text{sludge}$ ,

with  $r^2 = 0.975$ .)

estimated uncertainty of  $\pm 18\%$  for the emission measurements is at the same level as tracer gas based measurements reported from USA, which showed an uncertainty of 17% (Czepiel et al., 1996). This value was based on the square sum of all factors' uncertainty (RSS) for a methane plume 100-200 ppb above the background, excluding the effect of the tracer gas position. This included the tracer gas emission with an uncertainty of  $\pm 10\%$ , the tracer gas measurement with  $\pm 10\%$  and methane elevation in the plume with  $\pm 10\%$ . A corresponding evaluation of our study with a CH<sub>4</sub>plume at 100ppb and a N<sub>2</sub>O plume at 15ppb (comparable tracer gas measurement precision) would give an uncertainty of  $\pm 14\%$  (1 $\sigma$ /mean). Experiments, where two or more tracer gases have been released from the same area and used for estimating the calibration of the respective 'known' fluxes, have shown uncertainty levels (RSS) of 14% (Lamb et al., 1995) and 11%-21% (Mellqvist, 1999).

The methane production seemed stable over time, at least at Filborna, where most of the measurements were done (Fig. 2, Table 5). At Visby, we observed low ratios between measured production and potential production, and with the island of Gotland's special geology, lateral migration of LFG cannot be excluded. Such a process could have been further promoted by the additional cover applied in autumn 2002, between the last two measurements. No external methane source could be detected by the FTIR instrument measurements, so, if a lateral migration occurred, either the methane was completely oxidized leaving no isotope signal or the attenuation was so effective that emissions could not be detected above the background level.

The ratio between methane emitted to the atmosphere and total amounts of produced methane varied among the landfill sites within the range 20%–63%. Values for gas recovery were in a range 28%–78% during normal operation, and down to 14% at measurements done during periods of management problems. A study employing the tracer gas technique in the USA (Mosher et al., 1999) at five landfill sites showed similar values, with a ratio between methane emitted to the atmosphere and produced methane of 20%–50% (assuming 10% oxidation).

The mean gas recovery efficiency of 51% is lower than what has been reported by others, for example, 90% at a closed site in the USA (Mosher et al., 1999) and 69%–79% in Finland (Lohila et al., 2007), but does not support a default value as low as 20% suggested by IPCC (2006). The reported gas recovery of 34 kton methane in Sweden during 2002 (RVF 2003) means that 34/102 = 33% was utilised, which could be compared to the 51% efficiency that we observed. However, this 18% difference could easily be explained by the fact that only the 60 largest landfill sites had gas extraction systems installed.

Methane oxidation constituted only a small part of the total production according to our data, except for the closed landfills. For estimates of methane oxidation, the precision in the calculations is most commonly described as the standard deviation, without considering the uncertainty caused by the fractionation factor. Liptay et al. (1998) reported an estimated mean error of  $\pm 2\% - \pm 34\%$  for the precision in  $\delta^{13}$ C-measurements done at six landfills in the USA. When all parameters except  $\alpha$  vary. the Filborna samples are in line with this (-34%, +31%, 95%)confidence interval). The methane oxidation measurements (see Table 5), showed a variability of  $\pm 29\%$  ( $\pm 1\sigma$ ) in relation to estimated oxidation. Chanton et al. (1999) recorded a variability of <3%-23% in a landfill study in the USA comprising measurements done on 14 occasions, and their estimates were based on 11 samples for each subvalue as average compared to three in our study.

The effect of temperature was obvious for the active landfill sites, even though there were considerable differences among the sites concerning types of waste, cover materials, etc. Interestingly, both of the two closed landfills showed the highest ratios of oxidised methane, with maxima of 43% for Hagby and 37% for Visby, without any obvious correlation to temperature. However, this was based on only three observations. We must also be aware of the relatively large uncertainty in each estimate of the oxidation and the fact that soil temperature was only measured at one point at each site, and this value may not be representative for the conditions where the main part of the oxidation occurred. However, studies made by Czepiel et al. (1996) showed an optimum for oxidation at around 5–10 cm depth, similar to that also observed by Scheutz et al. (2003) and Christophersen et al. (2001).



Scheutz et al. (2003), who used the carbon isotope technique for measuring a closed landfill in France in September 2001 with a soil temperature of 22°C, arrived at an estimate of 40% oxidation, which is in level with the values we obtained for our two closed landfill sites, Hagby and Visby. The cover at the French site was constructed of a sandy soil with 2% organic matter, comparable to Hagby's 3.5% and Visby's 6%. In the same study, Scheutz et al. (2003) reported an oxidation of less than 4% in an area, which was not finally covered, but just amended with coarse sand and gravel. Supporting our data from the measurement in Sundsvall 9 March 2002, Christophersen et al. (2000) also observed considerable methane oxidation at low temperatures in winter. In the Sundsvall case, parts of the oxidation could have been either an effect of sunlight heating up parts of the surface or warm gas heating up the ground or a combination of these factors.

The total methane production levels deducted by the regressions (130 and 160 kg methane per ton MSW) corresponds to the normal mid-range of 120 kg methane per ton MSW reported by Themelis and Ulloa (2007). Our data also corresponds to the first national budget based on field data (Oonk and Boom, 1995) from 18 landfill sites in the Netherlands. Their estimate for 1993 ended at 282 Gg (364 formed, 51 recovered, 31 oxidised) with a range of uncertainty estimated at 170–405 Gg. This means a reduction of an earlier IPCC based budget by 25%, which is similar to our conclusion that the earlier IPCC model (IPCC, 2001), overestimated the methane emissions by around 41%.

The DOCf factor describes the fraction of the gas potential that is converted into methane. A comparison between measurement data and model calculations, using the IPCC model applied in the national climate gas reporting of methane from landfills, showed that the model overestimates the methane production in most cases. The earlier default value for DOCf at 0.77 (IPCC, 2001) was modified to 0.7 in the Swedish national report 2001 (Zuber et al., 2001), similar to what many other countries did at that time. The linear regression, which was calculated between landfilled DOC and our measured values of methane production gives a DOCf of 0.54, is comparable to the present default value of 0.5, now recommended by IPCC (IPCC, 2006, p. 3.13), but does not support the arguments for a DOCf-factor lower than 0.5 raised by Bogner and Matthews (2003)—given that DOC for the different waste fractions are not much altered.

We have not compared our field data with the latest IPCC model (IPCC 2006). As already stated in Section 1.3, data on waste were not collected accordingly, since the IPCC (2006) model requires a far more sophisticated differentiation for DOCf between waste types. The scientific basis for this differentiation is highly questionable, especially the regional defaults for these DOC values (IPCC 2006; table 2.3). It should also be mentioned that other models could be used (Scharff and Jacobs, 2006), which complicates international comparisons even further.

Concerning half-life time, contrasting results were observed for the methane production over time at the two closed landfills in our study. Visby showed a significant decrease (ca. 20% per year), whereas the methane production at Hagby showed no decrease at all. High amounts of building waste at Hagby (cf. Table 2), giving rise to lower degradation rates, is a likely explanation for the difference. The Swedish IPCC model (Zuber et al., 2001) used a half-life time of 7.5 yr, corresponding to a 9% annual decrease in methane production. However, there is a need for a more extensive database than the two landfill sites to evaluate this. The different k-factors (eq. 3), giving half-life times in the literature between 4 and 14 yr for LFG production (see Section 1.3. above), illustrate either that the conditions for LFG production are entirely different among countries, and/or that the assumptions are too rough. The latter case is supported by observations by Lagerkvist et al. (1997), who reported on the methane production in twelve test cells in three different Swedish landfills-after 5 yr, none of them had shown any decline, rather a more or less stable production during the experimental period. Some of the test cells in Brogborough, UK, also showed a continuous increase in the methane production, and even after 8 yr, no decline in the gas production of the six cells had occurred (Caine et al., 1999). Obviously, each landfill site has to be individually assessed to apply the most suitable model, and it's also necessary to integrate many landfills for a prediction at a national or regional level. Our data, especially the measurements made in Visby, show that data on gas recovery alone cannot be used to predict the half-life time, since the ratio between emissions and gas recovery (and oxidation) cannot be expected to be constant.

# 5. Conclusions—future research needs

The regulations in EU and in Sweden, together with a need for waste as a source of energy in incineration plants, has cut off most of the supply of organic waste materials to landfills. This will lead to a decreased gas production in the future. From 1994, when 1380 kton household waste was deposited in Sweden, the amounts have steadily decreased and during 2005 only 210 kton household waste was landfilled (RVF, 2006). However, considerable gas production will continue in the landfills for considerable time. How fast the decline will be, and how methane emissions will be affected during this decline is not known. We can expect lower gas production rates and lower gas quality in the future, which in turn will raise new demands on the functioning of gas extraction and flaring equipment, to avoid gas fluxes to the atmosphere or the risks with lateral migration of landfill gas (Gendebien et al., 1992; Christophersen et al., 2001). In this context, improved measurements, calculations and estimates of methane production, emissions and oxidation are of great interest. Improved methods will be important for verifying improvements in landfill management as well as getting correct data on how large the contribution is from each source on the national level, for the trade with greenhouse gases.

The precision for estimated methane production proved to be very good with our methods, down to  $\pm 4.2\%$ , which

enables trend studies and evaluation of improvements at landfill sites in the future. When it comes to absolute precision for production estimates, 95% confidence interval down to (-6.0%,+6.2%) were obtained. On occasions with high methane oxidation rates, the uncertainties will increase, especially if the recovered fraction is low. Thus, the best estimates of gas production will be achieved autumn–winter–spring, when the temperaturedependent methane oxidation is low.

Our data showed a strong correlation between deposited household waste and methane production. This was also true for the amounts of deposited DOC, but difficulties to judge on the DOC-content in the different waste fractions make this comparison more uncertain. For instance, a sludge factor at 5%-9% is most likely too low in the IPCC default values (ca. 25% measured at Filborna in this study and 27%-52% in Japan-Yamada et al. report cited in IPCC, 2006, p. 2.15). For improving the IPCC model, there is a need for an extension of measurements to include more landfills. The estimated DOCf would be improved by undertaking more measurements on landfill sites, that for long time have received almost constant (and known) waste amounts. To be able to calculate the half-life time for the methane formation process measurements from closed sites are needed. Also, with more measurements, more variables could be tested in the model. Furthermore, a better characterization of some waste fractions concerning degradability, especially the different types of industrial waste would add to a more comprehensive picture.

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