

The Swedish landfill methane emission project

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SUMMARY: Tracer technique, in combination with time resolved downwind plume concentration measurements by FTIR spectroscopy, has been applied for estimating total methane emissions from landfills in Sweden. Results from four landfills, constituting a set of different scales and topographies, are presented here.

1. INTRODUCTION

Atmospheric methane contributes to the greenhouse effect, and the concentration in the atmosphere increases globally by 0.6-0.8% per year (Houghton et al., 1995). The IPCC has estimated that more than 10%, 20-70 Tg year⁻¹, of the total anthropogenic methane emission originate from landfills (Prather et al., 1995). The organic carbon available in a landfill is biogenically degraded to CH₄ or CO₂, about 50% of each. Since CH₄ has a GWP (Global Warming Potential) that is 20 times stronger than CO₂, it is important to have a large fraction of the carbon to be emitted as CO₂. This can be achieved by applying an efficient gas recovery system, pumping the gas out for incineration, and/or by having an efficient bacterial methane oxidation in the top soil-layer of the landfill. In order to be able to validate the effect of measures taken to improve the performance of these systems, reliable methods for measuring the total emissions of CH₄ from a landfill are needed. Since 1997, a method based on tracer technique and time resolved concentration measurements by FTIR spectroscopy in the downwind plume of the landfill, has been developed and applied for estimating total landfill methane emissions in Sweden (Galle et al., 2001, Galle et al., 1999). After comparing several methods, it was decided to use the tracer gas technique for this project, which was initiated in 1999. The aim of the project is to improve the national budget of methane emissions from landfills in Sweden, as well as study the efficiency of the methane conversion systems at the different sites and supply a tool for validation of improvement measures. The methodology used is described, and the applicability of the method under different conditions is discussed, addressing parameters such as size, topography and meteorology. Preliminary measurement results from four different landfill sites are presented.

2. EXPERIMENTAL SECTION

2.1 Methane emission measurement

Fourier Transform Infrared Spectroscopy (FTIR) is used to obtain time resolved concentration measurements of methane in the downwind plume of the landfill. FTIR spectroscopy is an optical technique allowing a wide spectral region to be recorded simultaneously. Infrared light is transmitted over a long path in the gas under study, and an absorption spectrum is recorded. Thus a number of interesting gases can be analysed simultaneously in time, for instance CH_4 , N_2O , CO_2 , CO , H_2O and hydrocarbons. A long optical path is used to provide low detection limits, and typically sensitivities down to a few ppb are obtained. In this system a medium resolution (1 cm^{-1}) FTIR spectrometer is connected to an optical multiple-reflection gas cell with an adjustable pathlength, ranging from 9 to 107 meters. Normally a pathlength of 96 m is used, selected to optimise optical throughput and absorption levels. The system is built into a well-tempered and mechanically stabilised unit, and is located in a normal transport van. The recorded spectra are analysed by multiple-regression techniques, fitting synthetically derived calibration spectra of all present compounds (Griffith et al., 1996). CH_4 is analysed in the wavenumber region around 2950 cm^{-1} , and N_2O around 2200 cm^{-1} . Figure 1 shows a schematic view of the optical set-up.

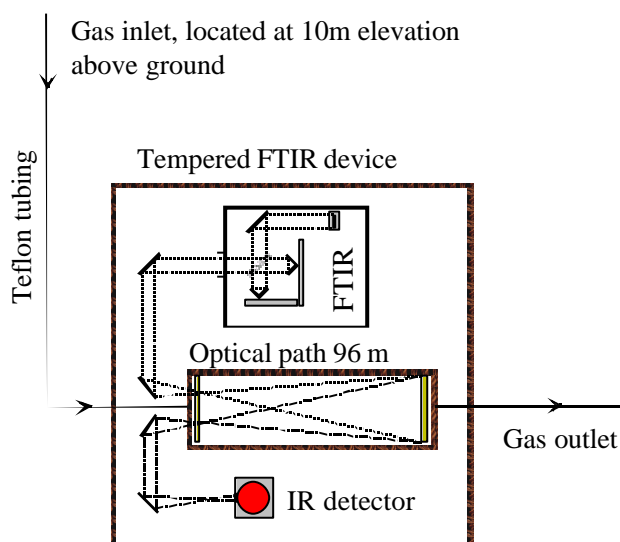


Figure 1. Schematic view of the experimental set-up.

The gas to be analysed is continuously pumped through the gas cell at a rate corresponding to one cell volume exchange per minute. Spectra are normally recorded averaging over one minute (corresponding to 13 added scans). In order to test the detection limit of the system at normal operation, consecutive 1-minute spectra were recorded for 5 hours measuring the same gas volume. The maximum deviation between two measurements (peak to peak) during a one hour period, which is the typical time for an emission measurement, was 2.7 ppb for methane and 0.7 ppb for the tracer used (N_2O). The spread of the measurements (Root Mean Square) over a one hour period was 0.59 ppb CH_4 and 0.16 ppb N_2O .

The optical system is accompanied by a meteorological system, consisting of one part that is stationary at the landfill, and one part that is located in the mobile measurement van. The stationary system is located at the top the landfill. Wind speed and wind direction are measured from a 5 m tripod mast, as well as air temperature, relative humidity and soil temperature (at 10 cm depth). All data are averaged over 1 minute, and saved to a battery-powered logger. On the

roof of the mobile van, a telescopic 10m mast is mounted. The gas inlet is located in the top of this mast in order to sample air from within the wind plume, and wind speed and wind direction at the measurement position are measured right beside the inlet. The sampled air is then transported through inert Teflon tubing to the gas cell. Other meteorological parameters measured are air temperature, relative humidity, barometric pressure, and pressure respectively temperature in the optical cell. The measurements are synchronised with the spectrum collection and stored with the spectra. Power is supplied either by the vehicle engine, or by a separate power generator.

The methodology used to couple the concentration measurements to an actual emission, is the *Time Correlation Tracer* method. A tracer, N₂O in this case, is released in a controlled way from the methane emitting area, by use of several point sources distributed over the landfill. The tracer will mix with the emitted methane in the landfill plume, and by time resolved analysis of the CH₄ and tracer concentrations far enough downwind the landfill, the emission can be derived. The part of the time series where the concentrations correlate, can be assumed to have its origin in the area where the tracer is released, and can be quantified using the known tracer flux

according to equation 1:
$$Q_{\text{CH}_4} = Q_{\text{Tracer}} \cdot \frac{C_{\text{CH}_4} \cdot M_{\text{CH}_4}}{C_{\text{Tracer}} \cdot M_{\text{Tracer}}} \quad (1)$$

Here C corresponds to mixing ratio and M to molecular weight. As the wind plume sweeps in and out over the measurement location, it can be judged how well the tracer release simulates the methane emission by looking both at the concentration correlation and the wind direction, and thus whether the total emission or only a fraction of it is simulated.

Depending on the size of the landfill 2-6 tracer point sources are used, each releasing at maximum 2.8 kg h⁻¹ of N₂O. The released amount of tracer is controlled by rotameters and integrating gas flow meters, and by weighing the gas tubes. The tracer release is remotely controlled (on and off). Also the distance to the measurement location is selected depending on the landfill size, and typically distances between 600m and 2400m are used. In order for the method to be applicable, a well-defined plume must exist and be reachable. That is, there should be wind speeds above 2 m s⁻¹ and some open terrain at the required measurement distance downwind the landfill.

2.2 Methane oxidation estimation

The methane oxidation rate is determined with the use of the isotope method, developed for landfills by Jeff Chanton and colleagues at University of Florida, USA (Liptay et al. 1998). The theory of this method relies on that methanotrophic bacteria discriminate ¹³C for ¹²C in methane, and thus methane emitted from the landfill surface will be enriched in ¹³C compared to methane in the anaerobic zone. In order to estimate the oxidation rate, knowledge is required about how large this discrimination is, i.e. the fractionation factor α_{ox} must be determined. The fractionation factor varies with soil type and temperature, and thus fresh soil samples from the different landfills are incubated with methane in laboratory studies. In the project described here samples for isotopic analyses are taken from

- The anaerobic zone (gas recovery system)
- The downwind landfill plume.
- The background
- Incubations (laboratory studies)

Sampling from the landfill plume is assumed to give the most representative average of the oxidation rate for the landfill, as this method includes integration over the landfill surface.

3. CHARACTERISATION OF THE LANDFILL SITES

In Figure 2 the geometry of the four landfill sites studied is schematically described, and the set-up of the tracer gas sources for the different measurement occasions is also shown.

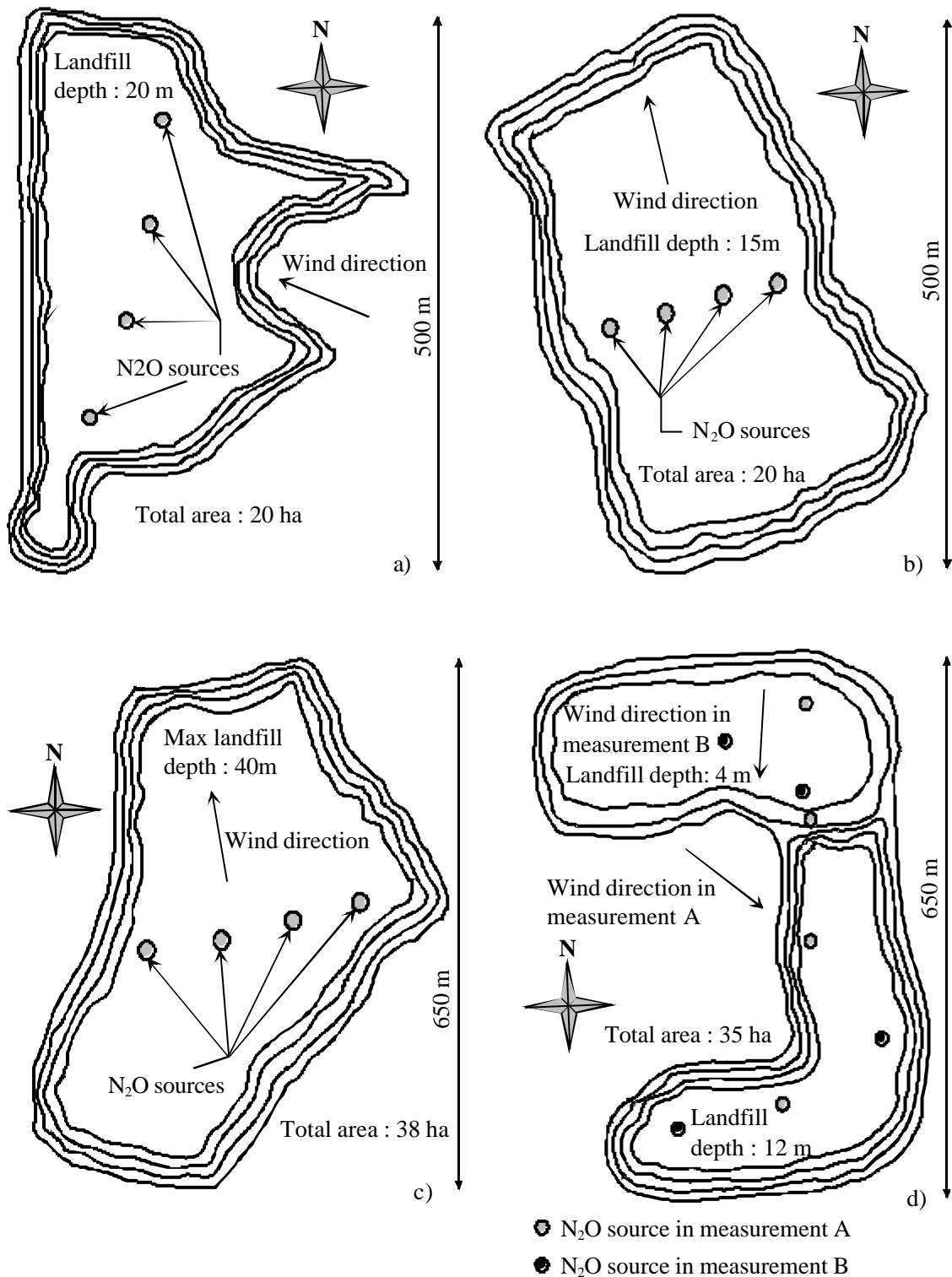


Figure 2. View over the different landfills and the tracer set-up at the different measurement occasions, a) Häljestorp (29th of March 2001), b) Hagby (20th of April 2001), c) Filborna (4th of April 2001) and d) Härlövs Ängar (12th of April 2001).

The sites studied in the project have been chosen to represent different aspects, such as: landfill size, waste characteristics (age, amount, type of waste), geographical location, gas extraction and management practises. The four landfills that are presented in this paper are all located in the southern half of Sweden, and they have been measured at during the period end of March to end of April 2001. All landfills are equipped with gas extraction systems. Table 1 displays parameters of interest for the different sites. The figures for the total amount of waste and the methane-producing fraction, involves a great deal of uncertainty, especially for the early years. From 1997 the different waste fractions put into Swedish landfills have been weighed (law enforcement), but some landfills have done this since the beginning of the 1990:s. Data have been supplied by inventory reports (Swedish Association of Waste Management, 1996) or by the landfill operators. In cases data is missing, estimates have been made using the total volume of waste and typical weights (Swedish Association of Waste Management, 1998).

Table 1. Landfill characterisation.

Landfill	Start	End	Total waste (10 ⁶ tonnes)	CH ₄ producing (10 ⁶ tonnes)	Household waste 1996-2000 (tonnes)	Gas recovery MWh ⁻¹ (1999)	Area (ha)
Filborna	1970	Active	11.5	8.3	297000	71706	38
Hagby	1968	1995	3	2	0	15693	20
Häljestorp	1975	Active	2	0.6	170000	10035	20
Härlövs Ängar	1959	Active	3.5	2.3	85000	18600	35

4. RESULTS AND DISCUSSION

The largest landfill in Sweden, according to methane production, is Filborna (Figure 2 (c)), with an area of 38 hectares. The topography is somewhat variable with different parts reaching different heights, but adding up to a hill formed shape with a maximum elevation of 40 m above the surroundings. The landfill is surrounded by flat agricultural land mixed with built-up areas. The measurement system was put up 2.2 km downwind the landfill (NNW), and measurements were carried out during two hours. The concentration time series of CH₄ and N₂O (with the ambient levels subtracted), along with the corresponding linear regression of CH₄ versus N₂O are shown in Figure 3. Due to the extension of the landfill, a large distance to the measurement location was required. This in turn demanded a large tracer release in order to obtain appropriate levels of N₂O at this point. Four tracer sources were evenly distributed on a line in the centre of the landfill, cross the wind direction. The total tracer release rate was 10.63 kgh⁻¹ of N₂O, giving a maximum signal of 10 ppb of N₂O at the measurement location (wind speed 6 m s⁻¹). It can be seen that CH₄ and N₂O vary synchronously in time. Observing that the slopes of the concentration curves coincided as the plume swept in and out of the location (from right to left), it could be assumed that the tracer release simulated the entire methane release. An estimate of the total landfill emission was thus obtained. From the slope of the regression line a methane emission of 308 kg h⁻¹ was calculated, and during the period of measurement the gas recovery rate was 700 kg h⁻¹. Depending on the meteorological conditions, it is estimated that an accuracy of 15-30% in the emission estimate is achievable (Galle et al. (2001)).

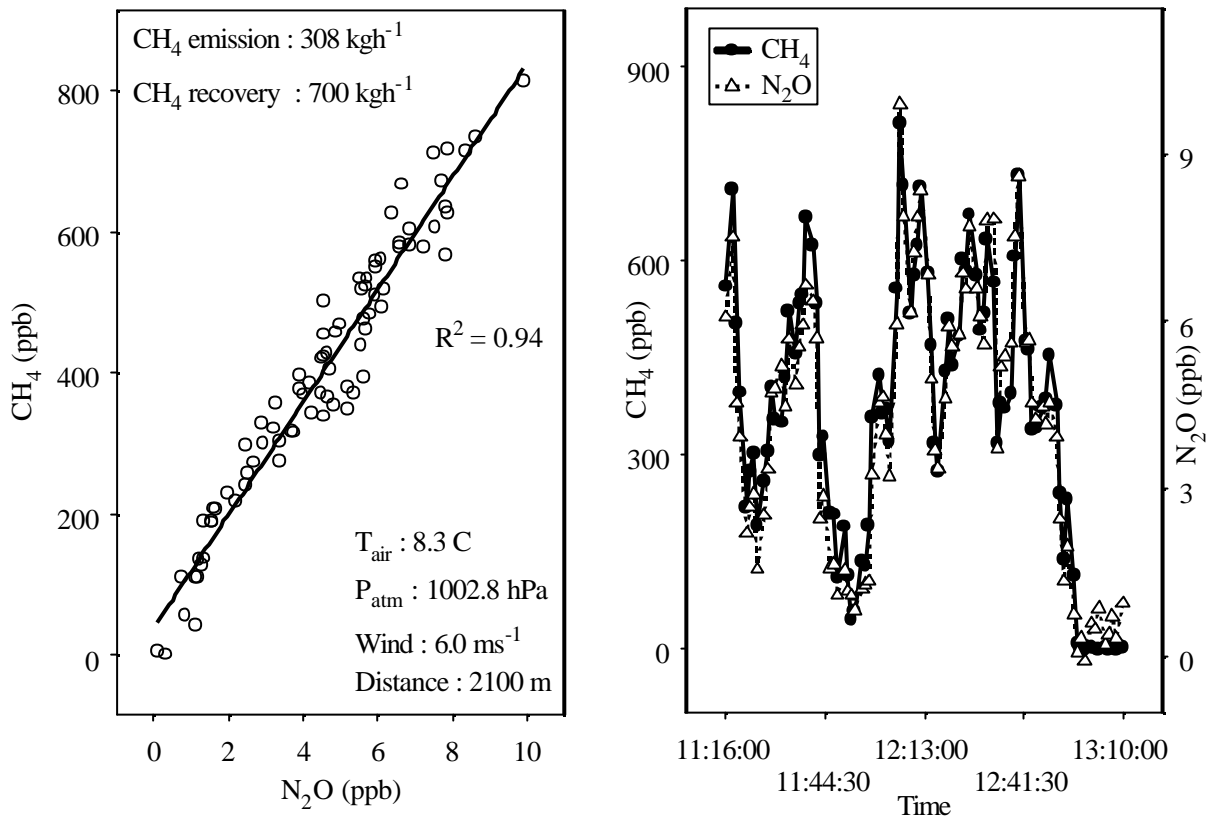


Figure 3. Emission measurement at the Filborna landfill on the 4th of April 2001.

A great benefit with the applied methodology is the possibility to judge on site, in real time, whether the measurement is successful or if any adjustments have to be made. This is exemplified by another measurement at the Filborna landfill, where the measurement system was put up at a distance of 1 km west of the landfill, facing its widest cross section. The concentration time series, along with the wind direction are shown in Figure 4. By observing the slopes as the plume crossed the measurement location, it was clear that the tracer release did not simulate the CH₄ release well, and the total emission could not be obtained unless the distance to the landfill was increased. Particularly the southern section of the landfill was not well covered by the tracer in this trial experiment.

Figure 5 displays the measurement results from the landfill Hårlövs Ängar, and the tracer set-up and a view of this landfill are shown in Figure 2 (d). Two measurements were carried out during the same day, between 11:45-12:45 (A, ESE of the landfill) respectively 17:05-18:05 (B, SSW of the landfill). The first measurement indicated a CH₄ emission of 142 kg h⁻¹, while the second measurement estimated the emission at 45 kg h⁻¹. In measurement B, CH₄ and N₂O correlated well with each other, and decreased synchronously when the plume swept in and out of the measurement location. This excludes the possibility that the emission was underestimated in measurement B.

The old northern part of the landfill, with a depth of 3-4 m, is likely to emit less methane than the newer southern section, with a depth of 10-12 metres. This is also indicated, comparing the obtained CH₄ levels from the northern and the southern edges of the landfill, as shown in Figure 5 (b).

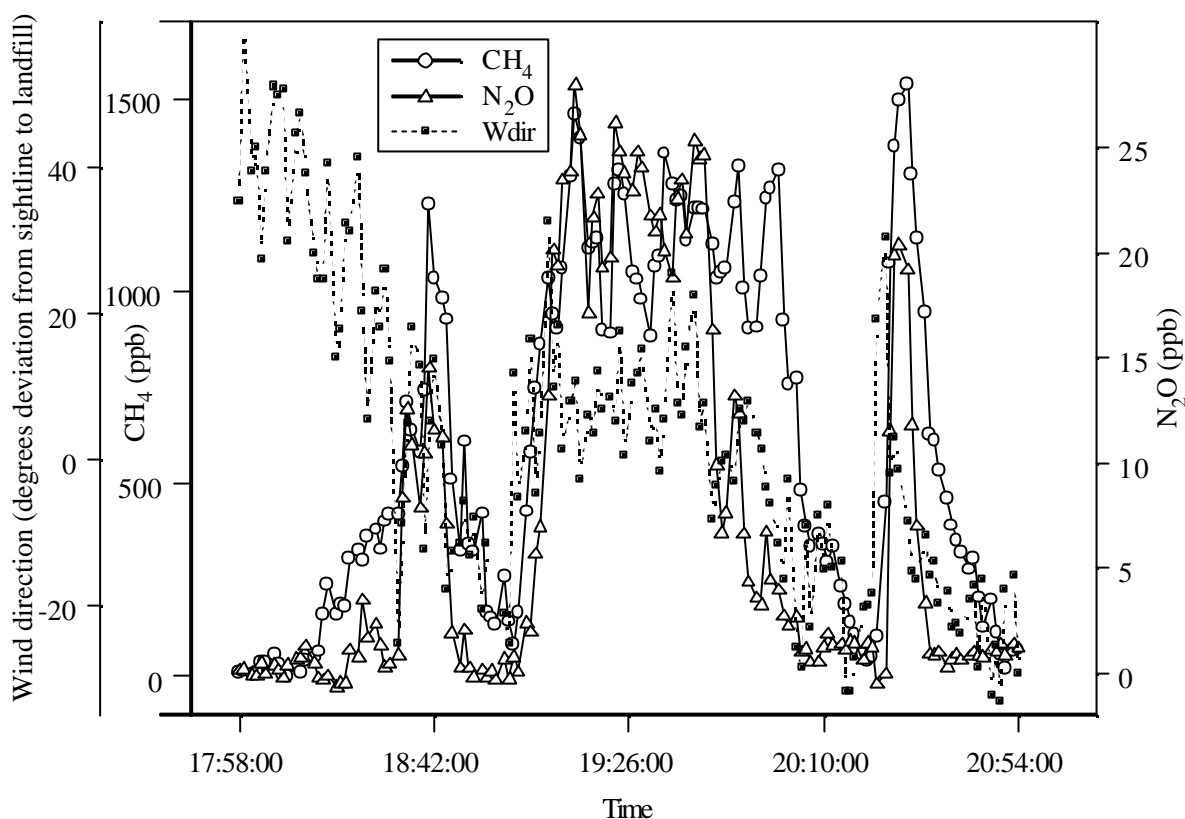


Figure 4. Measurement at the Filborna landfill on the 3rd of April 2001.

The tracer release was evenly distributed over the landfill in measurement A, and the observed N_2O concentrations were also symmetric around the centreline of the landfill. The regression plot of CH_4 versus N_2O should therefore show a distribution in the scatter points, corresponding to the two areas, having differing source strengths. The number of data points is not large enough to make this very clear, but an indication of a steeper slope corresponding to the southern, more active part, and a weaker slope corresponding to the old northern part can be seen. Nevertheless, observing the number of data points in the different wind direction segments, it is seen that the old and the new part have contributed about equally long time each to the measurement. The obtained emission for the two areas should therefore be weighted equally, calculating the total emission. This is equal to making the linear least squares regression to all data at once (which is done in Figure 5). The fact that the maximum level of CH_4 obtained in measurement A is about 2.7 times larger than in measurement B, despite comparable meteorological conditions and distances (i.e. similar dispersion), indicates that the observed emission difference originate in an actual emission decrease between the two occasions. The gas recovery system at this site on average delivered 117 kg h^{-1} , but according to the landfill operator there were problems with the recovery system during the day. The effect of a badly operating gas recovery system during measurement A, is the likely explanation to the deviation of 97 kg h^{-1} between the two emission estimates.

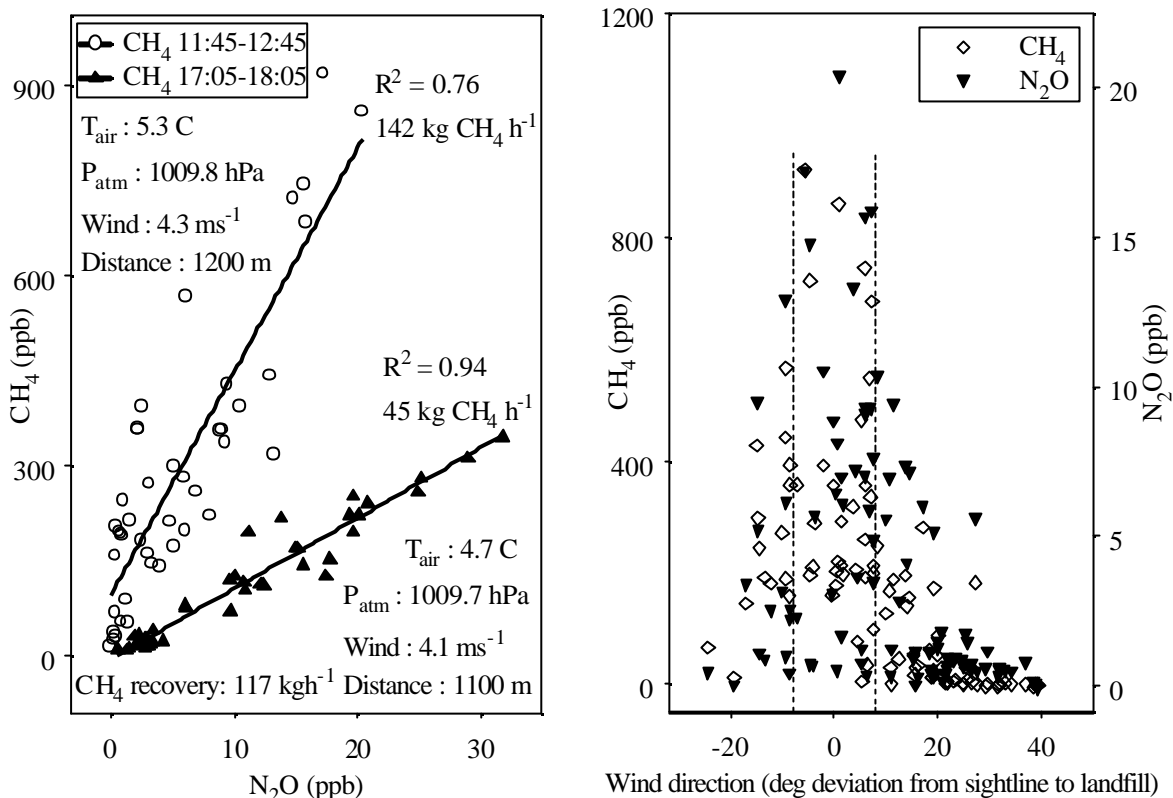


Figure 5. Measurement on the HÄrlövs Ängar landfill on the 12th of April 2001. The graph to the right corresponds to measurement B, and here the regions to the left respectively to the right of the dotted lines, correspond to the northern respectively the southern part of the landfill. The left graph includes the regression lines for both measurement A and B.

Using the Time Correlation Tracer method, the emission source is assumed to be fairly constant over the studied period. Deviations from this are encountered as larger scatter around the estimated ratio of CH₄ to N₂O. Still, making a linear least squares regression for the data gives the average ratio for the period. Another source of scatter is of course a poor simulation of the emission by the tracer. This can be due to either wrong area distribution of the tracer sources or a poor mixing due to weak winds. For wind speeds above 2 m s⁻¹ good mixing are normally observed. Generally, the uncertainty of the emission estimate due to the tracer source distribution, especially in the case of complex shapes and topography, is decreased if the distance to the measurement location is increased, making the landfill tend more and more to a point source.

Figure 6 displays results from the measurements at Hagby landfill (left) respectively Häljéstorp landfill (right). At Hagby, tracer gas was released over a period of 1.5 hours. In Figure 6 this period has been split up into three consecutive time periods, showing a successively increased emission over the period. During the whole measurement period the meteorological conditions were stable, and about the same maximum levels of nitrous oxide were detected in each time period, indicating similar dispersion. The observed increase in the CH₄ concentration over the periods thus can be assumed to reflect an increase in the emission. The estimated emission over the whole time period was 49 kg h⁻¹. Since the gas recovery rate was not available

at this short time resolution, it has not been verified whether variations in the recovery rate were responsible for the observed emission increase. The daily average for the gas recovery was 155 kg h^{-1} . The result of the measurement at Häljestorp shows a methane emission of 136 kg h^{-1} , which can be compared with a CH_4 recovery rate of 73 kg h^{-1} . At the time of measurement only about half of the methane producing area had been connected to the gas recovery system. This corresponds well with the poor gas recovery efficiency observed at this site compared to the others.

The Swedish landfill methane project also includes estimation of the methane oxidation in the landfill top covers, but at this time no results from this part are available to be presented.

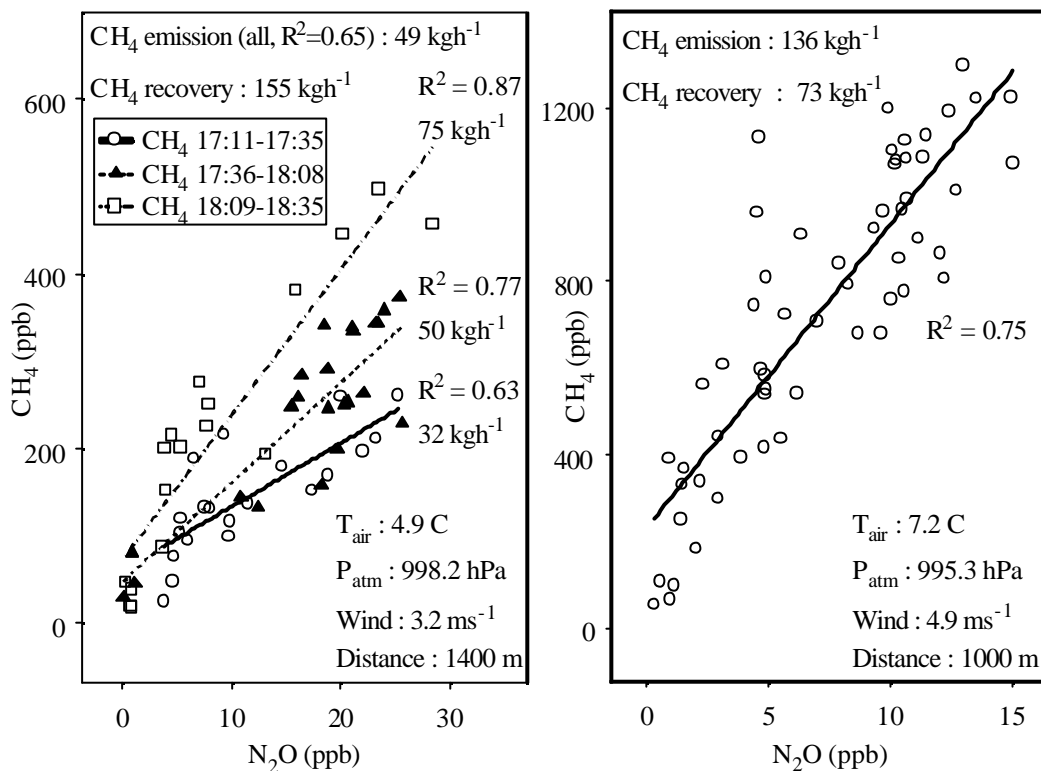


Figure 6. Measurement on the Hagby landfill (left) on the 20th of April respectively on the Häljestorp landfill (right) on the 29th of March 2001.

The estimates of the CH_4 emission at the different landfills, are summarised in Table 2, along with the daily averages of the methane recovery.

Table 2. Summary of the obtained methane emission estimates.

Landfill	Estimated emission ($\text{kg CH}_4 \text{ h}^{-1}$)	Gas recovery ($\text{kg CH}_4 \text{ h}^{-1}$)
Filborna	308	700
Hagby	49	155
Häljestorp	136	73
Härlövs Ängar	45	117

5. CONCLUSIONS

FTIR spectroscopy in combination with the Time Correlation Tracer method has proven to be an applicable method for total methane emission measurements at landfills of sizes ranging from small to large scale, provided that the surroundings downwind the landfill allows the wind plume to be reached. Dependent on the meteorological conditions each landfill required about 2-4 days, to obtain an emission estimate. Repeated measurements at the different sites will provide a useful statistic for estimating the gas recovery efficiency at the different landfill sites, and to improve the estimates of the yearly emissions of methane from Swedish landfills. In order to improve the time efficiency of the measurements, a strategy traversing the plume instead of letting the wind plume traverse the measurement location is considered (Czepiel et al., 1996). This facilitates studies of the edges of the landfill wind plume, getting information of the quality of the source simulation by the tracer release.

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REFERENCES

- Czepiel, P. et al., (1996), Landfill methane emissions measured by enclosure and atmospheric tracer methods, *Journal of Geophysical Research-Atmospheres* 101(D11), 16711-16719
- Galle, B. et al., (2001), Measurements of Methane emissions from landfills using a time correlation tracer method based on FTIR absorption spectroscopy, *Environmental Science and Technology*, 35, 21-25.
- Galle, B. et al., (1999), Measurement of methane emissions from landfills using FTIR spectroscopy, *Proceedings Sardinia 99, Seventh International Waste Management and Landfill Symposium*, CISA publisher, Cagliari, vol. IV, pp 47-54.
- Griffith, DWT. (1996), Synthetic calibration and quantitative analysis of gas-phase FT-IR spectra. *Applied Spectroscopy* 50(1), 59-70.
- Houghton, JT. et al., (1996), *Climate Change 1995, The Science of Climate Change, Contribution of WGI to the Second Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge, UK, Cambridge University Press.
- Liptay K, et al., (1998), Use of stable isotopes to determine methane oxidation in landfill cover soils. *Journal of Geophysical Research Atmospheres*, 103 (D7): 8243-8250.
- Prather, M. et al., (1995), *Radiative Forcing of Climate Change*. Houghton, JT and others. *Climate Change 1994. Radiative forcing of climate change and the evaluation of the IPCC IS92 emission scenarios*. Cambridge, UK, Cambridge University Press.
- Swedish Association of Waste Management, (1996), *RVF Rapport 1996:7*, (in Swedish)
- Swedish Association of Waste Management, (1998), *RVF Rapport 1998:9*, (in Swedish)