A comparison of methods for the measurement of methane emissions from municipal solid waste landfills for the purpose of evaluating their contribution to the greenhouse effect

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A COMPARISON OF METHODS FOR ESTIMATING METHANE EMISSIONS FROM MSW LANDFILLS

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INTRODUCTION

This study was funded and co-ordinated by the French Agence de l'Environnement et de la Maîtrise de l'Energie (Ademe) in the framework of the French research programme on the evolution of the climate and the atmosphere, with the specific aim of estimating the contribution to the greenhouse effect due to methane emitted by municipal solid waste landfills. The objective of the study was to assess and compare different methods for evaluating methane flux densities, with respect to the qualification of the methods in metrological terms, as well as their potential for practical implementation at landfill sites. This preliminary step, preceding the execution of direct measurements at selected sites, will enable researchers later on to validate existing emission models and establish by extrapolation an inventory of methane emissions covering all municipal solid waste landfills in France.

EQUIPMENT AND METHODS

Five methods were tested, in two sets of comparisons. Two localised methods were tested: accumulation chambers developed by the Institut National de l'Environnement et des Risques (INERIS) and by the Institut de Protection et de Sûreté Nucléaire (IPSN). Three atmospheric methods were tested: a mass balance technique and eddy correlation developed by the Institut National de la Recherche Agronomique (INRA) and a tracer-gas technique developed by IPSN. These methods were tested using two regulated-emissions devices installed by INERIS: a laboratory test-bed for the accumulation chamber methods, and an outdoors regulated-emissions site for the atmospheric methods.

Accumulation Chamber Methods

The INERIS method uses an external-recirculation chamber covering an area of 0.25 m^2 and with an enclosed volume of 30 litres (1). The internal atmosphere is renewed using an external pump with a flow rate on the order of 10 l/minute. The rate of methane enrichment of the recirculated mixture is monitored, and used to deduce the local methane flux of the covered surface. The methane flux derived outside the chamber is analysed using classical laboratory flame-ionisation chromatography. The chamber was validated beforehand in laboratory tests to ascertain the corrective factors to be applied to adjust for the influence of the measurement on the measured flux.

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The IPSN method uses a cylindrical accumulation chamber which also serves as the measurement chamber (2). It covers an area of 0.06 m^2 and has a usable volume of 19 litres. A fan placed inside the chamber ensures evenly-distributed filling throughout the accumulation phase. The chamber is equipped with instrumentation ensuring continual measurement of methane concentration, differential pressure between the interior and the exterior of the chamber, and temperature inside the chamber. The sensor is a semiconductor (TGS doped SnO_2, Figaro Engineering Inc., in an atmosphere desiccated by silica gel).

These two chambers were compared using a test-bed allowing regulated biogas emissions, built by INERIS. The bench consists of a 1m x 2m parallelogram-shaped bed of wet sand, roughly 30cm deep; a mixture of methane, carbon dioxide and water vapour is injected into the bottom of the bed under controlled conditions. The methane and carbon dioxide fluxes are regulated by mass flowmeters and distributed in the lower part of the bed by numerous pinhole-perforated tubes. The sand used in the bed and the emitted gas are heated to a temperature between 15 and 50°C by a heating pad placed in the lower part. On one side a fan is used to create air movements above the sand bed that simulate a wind of steady speed and direction. The $CH_4/(CO_2 + CH_4)$ mole ratio is maintained at a constant value equal to 60%. Methane flux densities ranging from 0.1 to 200 N ml / m^2 / minute can be obtained with this set-up. Six tests with different flux densities between 0.1 to 200 N ml / m^2 / minute. For two of the tests, d2w and d7w, a steady wind blowing at 4M/s was simulated.

Estimation of global flux on the basis of local measurements was not addressed in the framework of this comparison using these two non-integrating methods.

Atmospheric Methods

The eddy correlation method (INRA) relies on the principle that the flux density of a compound passing over a horizontal surface at a given time and place can be calculated as the product of the vertical wind speed and the concentration of the compound in question (3). This method requires measurement of the vertical wind speed, and of the compound concentration at the same place and moment in time with a frequency of at least 10 Hz. The experimental system is essentially composed of a three-dimensional sonic anemometer (Solent model, Gill) that measures wind components in an orthogonal system and air temperature at 20 Hz, and an adjustable laser diode spectrometer (Aerodyne Research Ltd.)

The mass balance method (INRA) relies on the principle that for a finite surface area, no matter what its characteristics, the flux emitted between the surface edge and the point of measurement is equal to the horizontal flow of the compound in question, integrated over the entire height of the boundary layer extending above the surface in question (3). The methane flux is estimated using measurement of wind speeds and methane concentrations at five heights in the lower atmosphere. Wind speed is measured using cupel anemometers (Cimel, Paris). Average methane concentrations are measured afterwards from air samples pumped into Teflon bags placed in rigid containers. All measurements and air sampling controls were carried out using a central data recording station (model CR10, Campbell Scientific Ltd.)

The tracer gas method (IPSN) consists in calculating the methane flux by measuring an atmospheric transfer coefficient (K) obtained using a tracer gas. Between an effluent source and a given point, K is the ratio between the integrated concentration of effluent at this point and the mass of effluent released (4). The method used consists in determining K by emission of a tracer gas under conditions that are as close as possible to those of methane emission, and in measuring simultaneously at the same point the concentrations of the tracer gas and of

methane. A constant mass flow, a known quantity, of the tracer gas sulphur hexafluoride SF_6 was injected into the methane emission network set up by INERIS. Sampling was done using independent gas sampling devices located at ground level around the site: most of the devices were placed downwind at about 4 metres from the last injection line, and one upwind of the site to determine the initial methane concentration. The tracer gas and methane concentration measurements were made at the site using two gas chromatography columns equipped respectively with a flame-ionisation detector for CH_4 and an electron-capture detector for SF_6.

For comparison of these three methods, an even, practically flat plot of ground with no background methane emissions from the soil was found in Etouy, in the Oise department north of Paris. The site was equipped with two sets of twenty 50-meter-long flexible tubes, arranged in parallel with a distance of 5 meters between each tube. The tubes were perforated by very thin metal pipes also 5 metres apart. Each tube was connected to a double central spine that ran the width of the plot along a median line. This double central spine emerged from a segment of single conduit through which all the flow passed. The gas emitted was a mixture of methane, nitrogen as vector gas and sulphur hexafluoride, in varying proportions. The emitted flow densities varied between 0.04 and 10.45 Nml/(m^2.min). A test repeating one density has already been carried out in order to ascertain the reproducibility of the methods.

RESULTS

Accumulation Chamber Methods

Table 1. Comparison of methane measurement results using accumulation chamber methods with flow densities actually emitted at test-bed

Tests	BENCH	IPSN Chamber		INERIS Chamber	
	average emitted flux density Nml/(m^2.min)	average emitted flux density Nml/(m^2.min)	uncertainty factor	average measured flux density Nml/(m^2.min)	uncertainty factor
d2	6.1	5	± 30%	6.0	± 5%
d2wind	6.1	11	± 30%	6.5	± 5%
d3	0.5	1	± 30%	0.5	± 5%
d4	180	71	± 30%	176.3	± 5%
d5	19	23	± 30%	19.2	± 5%
d6	20	16	± 30%	19.9	± 5%
d7	10	10	± 30%	9.6	± 5%
d7wind	10	11	± 30%	9.4	± 5%

The values for flow densities emitted by the test-bed are known within a relative uncertainty margin of ±5%. All the measurements determined by INERIS were correct. The results obtained with the IPSN chamber were less convincing. This chamber tended to underestimate high flux densities and overestimate low flux densities. In the most extreme cases the deviation from actual values reached 200%. Further test runs suggest that these deviations may be due to poor chamber design rather than to the sensor used.

Atmospheric Methods

Table 2. Comparison of measurement results using atmospheric methods with actually emitted flux densities

	EMITTED FLUX		INRA Eddy correlation method		INRA Mass balance method		IPSN Tracer gas method	
	Average density Nml/(m^2.min)	relative uncer-tainty (%)	average emitted flow density Nml/(m^2.min)	uncertainty Nml/(m^2.min)	average measured flow density Nml/(m^2.min)	uncertainty Nml/(m^2.min)	average measured flow density Nml/(m^2.min)	uncertainty Nml/(m^2.min)
A1	1.5	10%	1.0	0.1	1.5	0.8	1.7	0.5
A2	5.0	3%	2.6	0.2	3.8	1.0	5.6	1.2
A3	0.12	7%	0.04	0.03	1.4	1.5	0.3	0.2
A4	1.5	10%	0.6	0.1	2.0	1.2	1.7	0.5
A5	0.04	20%	0.02	0.01	0.8	1.0	0.14	0.09
A6	10.45	4%	5.8	0.5	5.0	1.0	9.9	2.3

The eddy correlation method systematically underestimated methane flux density by a factor of 1.5 to 3. This disappointing result has been attributed to a too-small test field and to the discrete level of methane emissions. The mass balance method correctly measured intermediate flux densities with a margin of error of less than 30%, but gave unsatisfactory results for very low values, that were overestimated by a factor of 10 to 20, and for very high values, that were underestimated by a factor of 2. Problems with the operation of the sampling system would appear to be at least partly responsible for the poor functioning of this method. The tracer gas method yielded good results for mid-range and high flux densities, with a margin of error of less than 20%. This method worked less well for low flow densities, that were overestimated by a factor of 2.5 to 3.5, even though the actual values were always within the uncertainty margin. This problem does not seem to be linked to the method itself, but to the difficulties encountered in measuring low methane concentrations.

DISCUSSION

This series of tests appears to establish the superiority of the INERIS method in the accumulation chamber method category, and that of the IPSN tracer gas method among the atmospheric methods. This conclusion must be nuanced, however. The final goal is to measure methane fluxes at actual municipal solid waste landfills, hence under conditions very different from the highly favourable conditions of the INERIS test-bed or the Etouy site. For instance, in the case of the INERIS accumulation chamber method, the test-bed measurements were made using a gas-phase chromatography column that would be replaced by a portable detector in the field. It is known that the biogas emitted at landfill sites has a complex composition. Care must therefore be taken to avoid interference problems when detecting methane with this type of detector. Regarding the IPSN tracer gas method, this method works well in principle if the methane and the tracer gas are emitted in identical fashion. This condition was fulfilled in Etouy, because the two gases were emitted through the same

network of injectors. But how can these conditions be respected at a real-life landfill, where methane fluxes will be unevenly distributed and not known prior to measurement? Inversely, the least effective methods in the comparison are not without relevance for real-life situations. The sensor mounted in the IPSN accumulation chamber is noteworthy for its low cost, light weight and low energy needs. Experimental conditions at the test site were probably not very favourable to the application of the INRA methods, particularly the eddy correlation method. The detector used for this latter technique performed remarkably well.

In conclusion, on-site experimentation now appears indispensable. Plans are to retain one accumulation chamber method and one atmospheric method for this further work, as the two types of method have not yet been compared directly. In light of the above results it seems natural to retain the INERIS chamber method and the IPSN tracer gas method. It would be interesting, however, to include the IPSN TGS sensor in the loop that INERIS will install on its accumulation chamber. Likewise, it would be interesting to ascertain if the adjustable laser diode detector used by INRA can improve measurement of low methane concentrations. If so, the effectiveness of the tracer gas method for low flux densities could be markedly improved by using the laser diode detector as a supplementary device.

NOTES

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