

DEVELOPMENT OF A SAMPLING PROTOCOL AND STANDARDS FOR UK LANDFILL GAS GENERATING SET EMISSIONS

Robert G Gregory, Andrew G Gillett

Land Quality Management Ltd
Nottingham
England

John H Blowes

Diesel Consult
Lancaster
England

ABSTRACT

Landfill gas contains a large number of trace components as well as methane. High temperature combustion, by flaring or utilisation of landfill gas, destroys a significant percentage of these compounds, potentially reducing the local and global environmental impacts. Conversely, combustion products are formed, some of which have local environmental impacts. There is, to date, little published data on the emissions of gas engines fuelled by landfill gas.

This research investigated landfill sites with power generation schemes utilising different designs of spark ignition and dual-fuel engines, fuelled by landfill gas of various compositions, to give a representative range of UK plant with which to objectively compare engine emissions. Research included the development of a standard, reliable and repeatable emission monitoring protocol based on recognised test methods. The aim was to produce a protocol that could realistically be utilised by operators when comparing emission levels to defined standards.

Detailed emissions testing was carried out on eight gas engines, and both bulk gas and trace gas compositions have been sampled in the supply gas and exhaust emissions in order to perform a "mass balance". Findings indicate the extent of environmental emissions of commonly regulated combustion products (NO_x, CO, SO_x, PCDDs/PCDFs, etc), up to 50 other trace component emissions (not reported in this paper), as well as bulk releases of CO₂, for comparison to future Environment Agency standards for landfill gas engines. Valuable information is also becoming available regarding typical engine performance tuning, which is currently based on attaining optimum thermal performance without the need to meet any specific emission limits. The knowledge accumulated will assist operators to determine which refinements to make to

achieve the limits that are likely to be set in the UK. Some options may have significant cost implications (the cost benefit work is being performed by LQM under a separate Agency Contract). Information generated by this project is intended to be used to supplement an Environment Agency project running in parallel with this project. Working alongside the regulator, we expect this research will help in the development of appropriate gas engine emission monitoring protocols and standards in future UK technical guidance. This paper reports interim data and our findings to date: the final report will be published in mid 2002.

INTRODUCTION

As part of an ongoing programme to revise UK technical guidance on landfill gas management, the Environment Agency for England and Wales (the Agency) issued in 1999 *Interim Internal Technical Guidance for Best Practice Flaring of Landfill Gas* (Environment Agency, 1999), but has not yet issued any comparable guidance on landfill gas engines. Nevertheless, there is increasing regulatory pressure on UK landfill gas utilisation plant operators to manage and/or reduce the local environmental impact of gas utilisation plant.

The Environment Agency, the licensing and pollution control Regulator in England and Wales, is in the process of carrying out a substantial amount of research on landfill gas engine and flare emissions, on the basis of which draft criteria for sampling and emissions standards are likely to be set. The drivers for these proposed standards are (1) the local human health and environmental impacts of landfill gas emissions, and (2) the impact on global warming of uncontrolled methane emissions from landfills.

There are, to date, little published data on the emissions, human exposure, or environmental impact of gas engines

fuelled by landfill gas, on which the issues surrounding landfill gas engine or flare emissions can be objectively debated. The Biogas Association commissioned LQM via the environmental body EB Nationwide to perform a series of trials to help quantify for the industry the range and magnitude of impacts from landfill gas fuelled generating sets so that these data could inform industry and regulators alike. This work is sponsored by landfill tax credits from Shanks, UK Waste (now Biffa) and Viridor, with seed funding from the three main gas engine manufacturers in the UK and seven gas utilisation companies. Subsequently the Agency commissioned Entec to perform a similar set of investigations, with the goal of developing a draft sampling protocol and possibly regulatory standards for certain combustion product emissions. These two projects (along with a third on flare emissions measurement (AEA Technology) and a fourth on the application of cost-effective gas cleanup technologies (LQM) have agreed to share concepts, data and information with the aim of developing a consistent view of emissions from landfill gas control and utilisation plant.

Currently, there are no statutory or non-statutory emission limits for landfill gas engines for the United Kingdom, but the *Interim Internal Technical Guidance for Best Practice Flaring of Landfill Gas* has been available for England and Wales since 1999 (Environment Agency, 1999). This provides recommended emission standards from flares stacks for CO, NO_x and unburned hydrocarbons, to ensure that the flare is operating correctly. These standards are reproduced in Table 1. In order to meet the emission limits it is recommended that the combustion air supply should be controlled in order to achieve a minimum flame temperature of 1000°C and retention time within this flame of 0.3 seconds. The guidance also proposes the replacement of low temperature open flares (except for emergency or test purposes for a maximum period of 6 months) for sites with existing waste management licences by enclosed designs by 2004, though this does not apply to old unlicensed landfills using open flares as they do not come under the control of the Environment Agency (Couth, 2001).

A recent study into the emissions from landfill gas energy recovery plant (Scott and Haigh, 1999) within the UK, investigated the emissions from four sites which included two landfill gas engines (one sampled twice) and a boiler fuelled by landfill gas. These data are reproduced as Table 2. Our experience of the types of engines measured prior to this 1999 study suggests that this early data are not representative of current UK practice, and should not be relied on as representative of current engine types or likely emissions from gas engines in operation today.

TABLE 1

**PROPOSED UK FLARE EMISSIONS STANDARDS
(ENVIRONMENT AGENCY, 1999)**

Substance	Flare emissions (mg/Nm ³ unless otherwise specified)
Reference O ₂ (%)	3
NO _x	150
CO	50
Unburnt hydrocarbons	10

REVIEW OF EUROPEAN STANDARDS

LQM reviewed emissions standards for flares and gas engines throughout Europe. It was clear from the findings of the review that the German and Swiss regulations are the most comprehensive in Europe for landfill gas engines, and other EC Countries appear to have applied a very much reduced set of criteria.

German Regulation

The Federal Immissions Control Act of 15 March 1974 provides the legislative framework for the air pollution program in Germany. The main objectives of this Act are the protection of the environment by meeting air quality standards to provide acceptable ground level concentrations and use of best-available technically feasible and appropriate control technology for the reduction of pollutant emissions. It allows the federal authorities, through the issuing of ordinances, to establish licensing requirements and set emission limits and control requirements for different types of facilities.

Three administrative regulations have also been issued under the Act. The most significant of these for industrial sources is the Technical Instructions on Air Quality Control (TA-Luft, 1986, as amended). These instructions set national standards for emission values and investigative procedures for the determination of these values.

The TA-Luft (1986) is the only applicable regulation that specifies parameters and limits for air quality. The latest amendments to the TA-Luft regulations were published on 1 March 1999. For the case of landfill gas (biogas) no changes to the original TA-Luft (1986) regulations are understood to have taken place. However, it is anticipated that amendments may be imposed in future regulations, following discussion with the European Commission (Wolfgang Stachowitz, personal communication, 2001).

TABLE 2
AVERAGED EMISSIONS DATA FROM A RANGE OF LANDFILL GAS UTILISATION PLANT
(AFTER SCOTT AND HAIGH, 1999)

Parameter	Site 1	Site 1 After Maintenance Service	Site 2	Site 3
Plant type	Internal Combustion	Internal Combustion	Internal Combustion	Boiler
Total Particulate Matter	1.54 (1.39 - 1.78) ¹	0.50 (0.44 - 0.55)	55.70 (43.1 - 63.2)	0.18 (0.07 - 0.25)
Heavy Metals	0.02 (0.00 - 0.025)	0.03 (0.02 - 0.04)	0.54 (0.07 - 0.90)	0.04 (0.08 - 0.04)
Total NMVOC	220 (180 - 340)	160 (145 - 150)	74 (120 - 68)	1.7 (0 - 5)
PCDD & PCDF (ng/Nm³ TEQ)²	0.017	<0.001	0.014	<0.001
PAHs²	0.009	0.007	0.025	0.001
Hydrogen Chloride	3.5 (1.7 - 6.5)	5.9 (5.8 - 6.1)	1.1 (0.2 - 2.50)	8.7 (3.5 - 13.5)
Hydrogen Fluoride	6.2 (3.1 - 22.7)	1.25 (0.2 - 1.7)	0.38 (0.0 - 1.2)	0.49 (0.0 - 1.0)
Carbon Monoxide	262 (245 - 287)	277 (219 - 359)	336.8 (309 - 386)	0.64 (0.0 - 2.5)
Oxides of Nitrogen (NO_x)	551 (479 - 823)	3100 (2787 - 3397)	296 (276 - 314)	15.7 (11.7 - 18.2)
Oxides of Sulfur	18.4 (4.7 - 48.8)	10.5 (0 - 44.0)	5 (1.1 - 13.2)	1.1 (0.0 - 7.2)
Nitric Acid	41.5 (33.1 - 51.0)	6.4 (5.6 - 7.0)	30 (27.6 - 7.0)	1.2 (0.0 - 3.7)
Phosphates²	<0.1	<0.1	<0.1	<0.1

Notes: All data expressed in mg/Nm³ for reference conditions of 15% oxygen content, 273K, 101.3 kPa and dry gas unless otherwise stated; ¹Mean value quoted where a range of values were obtained from different measurement techniques (range given in brackets); ²Single measurement per site obtained (no range given); NMVOC, Non-methane volatile organic compounds; PCDD, PCDF, polychlorinated dibenzodioxins and polychlorinated dibenzofurans respectively; PAHs, polycyclic aromatic hydrocarbons; TEQ, Toxicity Equivalents.

The TA-Luft regulations are not legally compulsory for German landfill operators, rather they act as a guideline for plant permitting by authorities. There are no more specific regulations in the Federal Immissions Control Act. The Ta Siedlungsabfall (Technical Guideline on MSW Management) specifies operation of landfills and gas collection, but no air pollution limits (Ivo Mersiowsky, personal communication, 2001).

Specific emission limits exist within the TA-Luft (1986) for the different types of facilities that may combust landfill gas:

- Furnaces for gaseous fuels with firing thermal capacity < 100 MW (emission limits considered valid for flares, Wolfgang Stachowitz, personal communication, 2001);
- Internal combustion engines; and
- Gas turbines.

These facilities are defined according to the Annex of the Ordinance on Facilities Subject to a Licence of July 24, 1985. Where the emission limits presented within Table 3 are not derived from the relevant facility regulation limits a footnote indicates any relevant conditions applicable to the stated limit.

The TA-Luft (1986) contains limits for a range of inorganic dust particles, grouped into the classes as indicated in Table 4 (Part III B of the MAK-Value-List).

Section 2.3 of the TA-Luft (1986) also presents emission standards for Part III A1 and A2 MAK-Value-List carcinogenic substances in their respirable forms (Table 5). If substances of more than class are present then the mass concentration in the waste gas must not exceed the total of the highest class limit.

TABLE 3
EMISSION LIMITS (mg/Nm³) FOR FACILITIES COMBUSTING LANDFILL GAS (TA-LUFT, 1986)

Substance	Furnace/Flare ¹ (gaseous fuel)	Internal Combustion Engines	Gas Turbines
Reference O ₂ (%)	3	5	15
Dust / Particulates	5	50 / 150 ²	2 / 4 ³ (Soot value) ⁴
SO _x (SO ₂ + SO ₃)	35	500 ⁵	500 ⁵
NO _x (NO + NO ₂)	200	500 (4-stroke) 800 (2-stroke) ⁶	300 / 350 ²
CO	100	650	100
HCl		30 ⁷	
HF		5 ⁸	
H ₂ S		5 ⁷	
NH ₃		-	
Organo S		-	
Cd		0.2 ⁹	
Total metals		-	
Organic C (NMVOCs in Table 5 below)		20 (Class I) ¹⁰ 100 (Class II) ¹¹ 150 (Class III) ¹²	

Notes: ¹ Furnace standards are used for flares in Germany (Wolfgang Stachowitz, personal communication, 2001). ² at a mass flow of >0.5 / <0.5 kg/h, respectively. ³ waste gas volume flow of ≥60 000 / <60 000 m³/h. ⁴ Soot number is the degree of blackening on a filter paper caused by flue gases (according to the Bacharach method) with 10 degrees from 0 to 9. ⁵ at a mass flow ≥ 5 kg/h. ⁶ engines without compression ignition. ⁷ at a mass flow of ≥ 0.3 kg/h. ⁸ at a mass flow of ≥ 50 g/h. ⁹ at a mass flow of ≥ 1 g/h. ¹⁰ at a mass flow of ≥ 0.1 kg/h. ¹¹ at a mass flow of ≥ 2 kg/h. ¹² at a mass flow of ≥ 3 kg/h.

TABLE 4
EMISSION LIMITS FOR INORGANIC DUST PARTICLES (SECTION 3.1.4, TA-LUFT, 1986)

Class	Element & compounds of	Mass flow (g/h)	Limit (mg/Nm ³)
I	Cadmium, mercury & thallium	≥ 1	0.2
II	Arsenic, cobalt, nickel, selenium & tellurium	≥ 5	1
III	Antimony, lead, chromium, cyanides, fluorides, copper, manganese, platinum, palladium, rhodium, vanadium and tin	≥ 25	5

TABLE 5
EMISSION LIMITS FOR CARCINOGENIC SUBSTANCES (SECTION 2.3, TA-LUFT, 1986)

Class	Element & compounds of	Mass flow (g/h)	Limit (mg/Nm ³)
I	Asbestos as fine dust; benzo(a)pyrene; beryllium and beryllium compounds; dibenz(a,h)anthracene; 2-naphthylamine.	≥ 0.5	0.1
II	Arsenic trioxide/pentoxide, arsenious acid & salts, arsenic acids/salts; chromium (VI) compounds: calcium chromate, chromium (III) chromate, strontium chromate, zinc chromate; cobalt (dusts/aerosols of cobalt metal and salts of low solubility), 3,3-dichlorobenzidine; dimethyl sulphate; ethyleneamine; nickel (dusts/aerosols of nickel metal, nickel sulfide and pyritiferous ores, nickel oxide & nickel carbonate, nickel tetracarbonyl)	≥ 5	1
III	Acrylonitrile; benzene; 1,3-butadiene; 1-Chloro-2,3-epoxypropane (epichlorohydrin); 1,2-dibromomethane; 1,2-epoxypropane; ethylene oxide; hydrazine; vinyl chloride	≥ 25	5

The Institution of the Environment of Hessen (Germany) recommend for the use of raw gas within combustion units at temperatures exceeding 1200 °C with retention times, within the high-temperature zone, of at least 0.3 seconds that the total chlorine and total fluorine should be less than 200 and 50 mg/Nm³ respectively. These standards were first introduced in 1989, but since 1999 they have no longer applied to the landfill gas industry (Wolfgang Stachowitz, personal communication, 2001).

Swiss Regulation

The most important piece of environmental legislation in relation to regulating landfill gas flare or engine emissions is the Environmental Protection Law (EPL) of 7 October 1983 which came into force on 1 July 1997 (EPL, 1997). The EPL (1997) provides for the application of emissions limits to combustion installations as laid down in the Luftreinhalte-Verordnung (LRV) or Ordinance on Air Pollution Control (OAPC, 1998). The OAPC contains preventive emission limits for combustion installations and industrial sites in relation to the impact thresholds established for the leading air pollutants. If the impact thresholds are respected then any harmful or irritating effects are unlikely to arise.

The responsibility for implementing the OAPC lies with the cantons and communes, who may pass their own environmental laws and regulations providing it does not contradict federal law. The cantons can issue retrofitting orders to ensure compliance with emission limits, monitor air pollution levels within their territory and draw up action plans and proposals to the Federal Council to comply with the impact thresholds. The Swiss government set up the Interdepartmental Working Group on Air (IDA-Air) in 1994 to co-ordinate the response to the OAPC, which has now been completed leading to its official dissolution in 2000 (Richard Ballaman, personal communication, 2001). The minimum requirements for reducing nationwide emissions of SO₂, NO_x, NH₃, particulates (PM₁₀) and VOCs have been laid down in the Air Pollution Control Strategy (APCS, 1999).

General preventive emission limits exist within the OAPC at Annex 1 for the control of emissions from stationary sources. These general limits are applicable to the combustion of landfill gas subject to the additional provisions laid down for:

- Specific waste installations, including installations incinerating municipal and special waste, in which the definition of municipal waste includes gases from waste as landfill gas (i.e. flares) for which the total chlorine and fluorine content is greater than 50mg/Nm³; and

- Specific installations (Annex 2), including stationary internal combustion engines and gas turbines that may combust landfill gas.

The applicable limits under the OAPC (1998) for the various installations capable of combusting landfill gas are presented in Table 6. Generally, the emission limits and classification of organic and inorganic substances as dust particles or in a gaseous form and the carcinogens are the similar to those listed in the TA-Luft (1986).

Discussion of limits applied in Europe

The emission limits for dust/particulates are generally higher for engines compared to flares, with a consistent limit of 50 mg/Nm³ applied. The TA-Luft (1986) set a less stringent limit for engines which have a mass flow less than 0.5 kg/h, of 150 mg/Nm³. The TA-Luft (1986) limit of 500 mg/Nm³ for SO_x is twice that stated within the Swiss regulations. In addition, the higher German limit for SO_x is defined for engines with exhaust gas mass flow rates ≥ 5 kg/h whereas the more stringent Swiss limit applies to engines with an exhaust gas mass flow rate ≥ 2.5 kg/h. It would be expected that more stringent limits should apply to engines with the higher SO_x mass flow rates.

The TA-Luft (1986) make a distinction between engines in setting NO_x emission limits, with limits of 500 and 800 mg/Nm³ for 4 and 2-stroke engines, respectively. The Swiss limit is again more stringent at 400 mg/Nm³ with the additional requirement of the inlet gas to have a total chlorine and fluorine content < 50mg/Nm³.

There is general consensus across Europe for CO, with an emission limit of 650 mg/Nm³. There is agreement of the emission limits for the acid forming gases of HCl, HF and H₂S with emission limits of 30, 5 and 5 mg/Nm³, respectively. Cadmium emissions limits are set at 0.1 mg/Nm³ by the Swiss regulations, half of that stated within the TA-Luft (1986).

Only the Swiss regulations set out engine emission limits for NH₃ and organo-sulfur compounds (carbon disulfide), with limits of 30 and 100 mg/Nm³, respectively. Both the TA-Luft (1986) and OAPC (1998) set separate emission limits for organic C substances dependent on their relative carcinogenic properties and mass flow rates, of 20, 100 and 150 mg/Nm³ for Class I, II and III carcinogens, respectively.

With these limits being applied in Europe, the UK has decided to perform a series of emissions tests to ascertain what practical emissions standards and routine sampling methods might be applied.

TABLE 6
EMISSION LIMITS (mg/Nm³) FOR FACILITIES CAPABLE OF COMBUSTING LANDFILL GAS (OAPC, 1998)

Substance	Waste installations (Flare)	Internal Combustion Engines	Gas Turbine
Reference O ₂ (%)	3	5	15
Dust / particulates	10	50	2 / 4 ¹ (Soot value)
SO _x (SO ₂ + SO ₃)	50	250 ²	120 ³
NO _x (NO + NO ₂)	80 ⁴	400 ⁵	50 / 150 ⁶
CO	50	650	120 / 240 ⁷
HCl	20		30 ⁸
HF	2		5 ⁹
H ₂ S	-		5 ¹⁰
NH ₃	5		30 ¹¹
Organo S	-		100 ¹²
Cd	0.1		0.1 ¹³
Total metals	1		-
Organic C (NMVOCs in Table 5 above)	20		20 (Class I) ¹⁴ 100 (Class II) ¹⁵ 150 (Class III) ¹⁶

Notes: ¹ heat input ≥ 20 MW / < 20 MW. ² at a mass flow ≥ 2.5 kg/h. ³ at a mass flow ≥ 2.5 kg/h. ⁴ at a mass flow ≥ 2.5 kg/h. ⁵ emission limit valid providing inlet gas has total chlorine and fluorine content < 50 mg/Nm³. ⁶ heat input ≥ 40 MW / < 40 MW. ⁷ heat input ≥ 40 MW / < 40 MW. ⁸ at a mass flow of ≥ 0.3 kg/h. ⁹ at a mass flow of ≥ 50 g/h. ¹⁰ at a mass flow of ≥ 50 g/h. ¹¹ at a mass flow of ≥ 0.3 kg/h. ¹² Organo sulfur listed is carbon disulfide, at a mass flow of ≥ 2.0 kg/h. ¹³ at a mass flow of ≥ 0.5 kg/h. ¹⁴ at a mass flow of ≥ 0.1 kg/h. ¹⁵ at a mass flow of ≥ 2 kg/h. ¹⁶ at a mass flow of ≥ 3 kg/h.

SAMPLING AND ANALYSIS

LQM set about developing a robust methodology for sampling and analysis of a large suite of bulk and trace components in the landfill gas supplied to the gas engines, and another suite of post-combustion exhaust gases, which could potentially be measured either at the exhaust manifold, post-turbocharger or post-silencer as part of a routine method for landfill gas engine operators to fulfil future emissions measurement requirements.

Sampling methods were required to give robust analytical results. No standard sample location points are fitted on the exhausts of the majority of UK plant, and so it was decided to design a sampling sleeve or “boot” which could be employed on the three main engine types employed in the UK with minimal modification.

The sampling sleeve, built from stainless steel, was designed to extend vertically the existing stubby exhaust sleeves of the engines studied so as to allow concomitant sampling of several species, and to facilitate isokinetic sampling in accordance with the British Standard BS6069. The sleeve had two 4 inch and two 2 inch BSP sampling ports fitted. The 4 inch ports were drilled at 90° to each other, 5 id from the inlet and 2 id from the final exhaust. The 2 inch ports were mounted below and offset 45° from the 4 inch ports, midway between the two 4 inch ports. Access and lateral support for the sleeve was normally

achieved by scaffolding. A schematic installation layout is shown in Figure 1. Figures 2 – 3 show the sampling sleeve and various sampling trains in use at a number of sites.

The supply gas was sampled from various points as close to the gas engine as practicable, and subsequent to any primary or secondary gas cleanup.

Analytical methods were chosen on the basis of reproducibility and accuracy. All samples were collected by our main subcontract laboratory, CPL Laboratories of Chesterfield, and analyses made by CPL, SAL, CERAM Research and Harwell Scientifics.

Landfill Supply Gas

Tables 7 and 8 summarise the methods employed for sampling and analysis of the supply gas.

Bulk Gases: analysis was determined in accordance with CPL “in-house” procedure, AM-001:1994 Pt 4 and involves thermal conductivity gas chromatography (GC/TCD). This procedure is UKAS (United Kingdom Accreditation Service) accredited for landfill gas analysis.

Total Sulfur, Chlorine and Fluorine: this was determined by a modified BS 3156 procedure “Methods for the Sampling and Analysis of Fuel Gases” using a microburner arrangement.

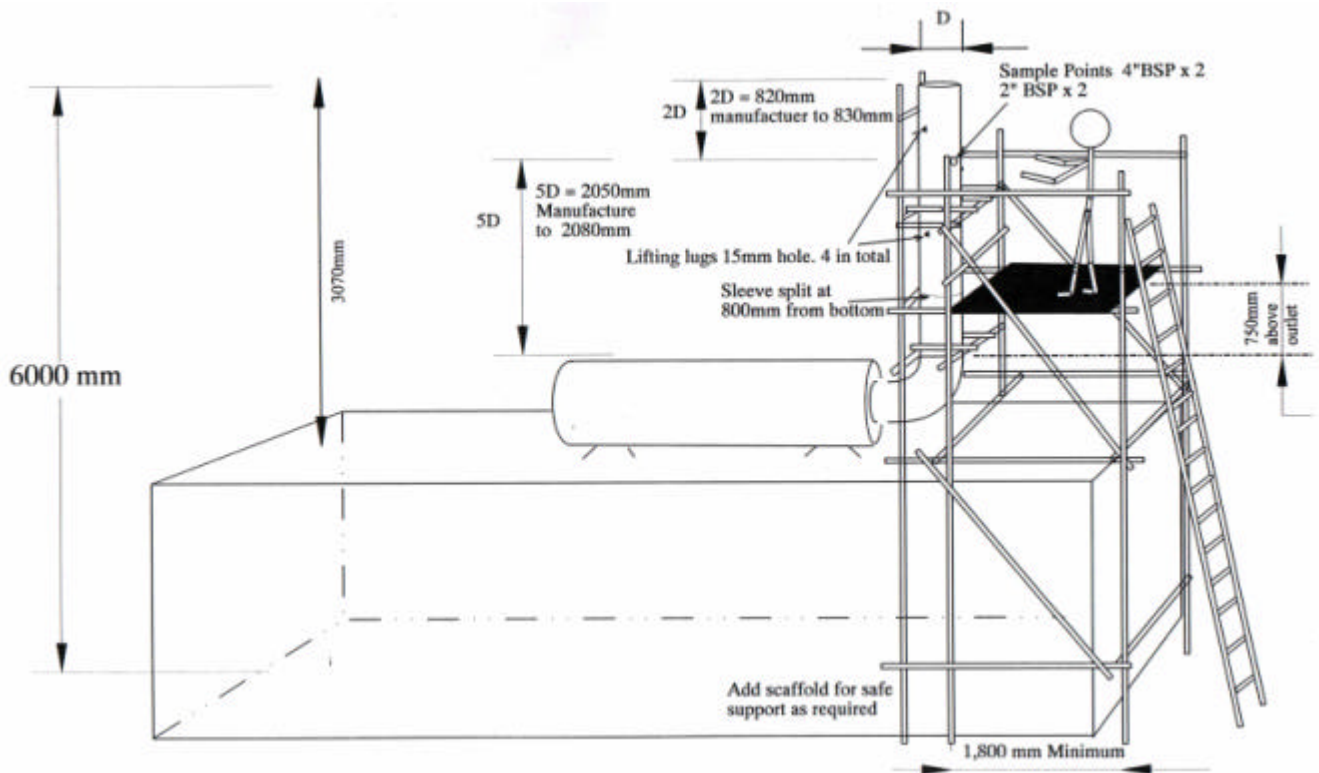


FIGURE 1. INDICATIVE LAYOUT OF THE SAMPLING SLEEVE FITTED TO A CONTAINERISED ENGINE UNIT, AND THE REQUIRED SCAFFOLDING TO ACCESS THE SAMPLING PORTS. ALL DIMENSIONS ARE NOMINAL AND PROVIDED FOR INDICATION ONLY.

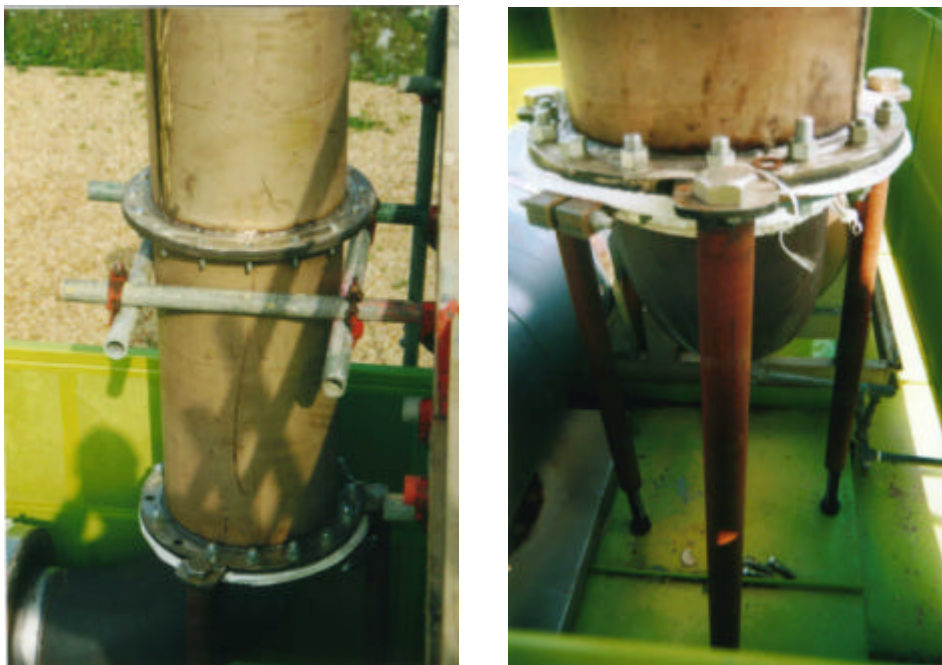


FIGURE 2. VIEWS OF INSTALLED SAMPLING SLEEVE SHOWING FIXING OF LOWER SLEEVE TO SCAFFOLDING PLATFORM AND USE OF SUPPORTING LEGS



FIGURE 3. SAMPLING PLATFORM INSTALLED ACROSS A CONTAINERISED GENERATING SET, AND CLOSE-UP OF PARTICULATE SAMPLING EQUIPMENT INSTALLED ON SCAFFOLDING

**TABLE 7
SUMMARY OF SAMPLING PROCEDURES FOR INLET GASES**

Determinand	Sampling method	Procedure No.	UKAS Ref No.
Methane	Gresham tubes	AM 001 pt 4	1618
Oxygen	Gresham tubes	AM 001 pt 4	1618
Carbon dioxide	Gresham tubes	AM 001 pt 4	1618
Hydrogen	Gresham tubes	AM 001 pt 4	1618
Total sulfur	Based on BS 3156	AM 001 pt 1	1618
Total fluorine	Based on BS 3156	AM 001 pt 1	1618
Total chlorine	Based on BS 3156	AM 001 pt 1	1618
Moisture	Silica gel	AM 001 pt 3	1618
Organosulfur	Teflon bags	Not applicable	Not applicable
Hydrogen sulfide	Teflon bags	Not applicable	Not applicable
Organosilicon	XAD Sorbent tubes	Not applicable	Not applicable
Carbon monoxide	Electrochemical cell and Gresham tubes	Not applicable	Not applicable
VOCs (speciated)	Gresham and sorbent tubes	AM2010/2011	Not applicable
VOCs (aldehydes)	Treated XAD-2 sorbent tubes	Not applicable	Not applicable

**TABLE 8
SUMMARY OF ANALYTICAL PROCEDURES FOR INLET GASES**

Determinand	Analytical method	Analysis house	UKAS Ref No.	Sample location	Archive period
Methane	GC/TCD	CPL	1618	Not applicable	Not applicable
Oxygen	GC/TCD	CPL	1618	Not applicable	Not applicable
Carbon dioxide	GC/TCD	CPL	1618	Not applicable	Not applicable
Hydrogen	GC/TCD	CPL	1618	Not applicable	Not applicable
Total sulfur	IC	CPL	1618	CPL	3m
Total fluorine	ISE	CPL	1618	CPL	3m
Total chlorine	IC	CPL	1618	CPL	3m
Moisture	Gravimetric	CPL	1618	Not applicable	Not applicable
Organosulfur	GC/MS	CERAM	0013	Not applicable	Not applicable
Hydrogen sulfide	GC/FPD	CERAM	0013	Not applicable	Not applicable
Organosilicon	GC/MS	SAL	Not applicable	SAL	3m
Carbon monoxide	IR	CPL	Not applicable	Not applicable	Not applicable
VOCs (speciated)	GC/MS	Harwell Sci.	Not applicable	Not applicable	Not applicable
VOCs (aldehydes)	GC/FID	SAL	Not applicable	Not applicable	Not applicable

Analysis of the products of combustion was subsequently effected by ion chromatography. This method is a documented CPL “in-house” procedure, AM-001:1994 Pt 1 and is UKAS accredited for landfill gas determinations.

Hydrogen Sulfide: this was determined by extraction into a suitable sampling vessel prior to analysis by gas chromatography incorporating a flame photometric detection unit (GC/FPD) at a sub-contract laboratory (CERAM Research, Stoke-on-Trent).

Volatile Organic Compounds (VOC): VOC content of the gas was established via two ex-situ techniques. Firstly, low boiling range compounds were analysed by packed column gas chromatography using flame ionisation detection following sampling into Gresham tubes. Secondly, the relatively less volatile compounds were identified by either:

- sampling onto “mixed-bed” sorbent material (including Carboxen 1000), prior to analysis by gas chromatography with mass spectrometry (GC/MS). The sub-contract laboratory, Harwell Scientifics, analysed for specific LQM nominated compounds considered to be potentially present in landfill gas (not reported in this paper).
- Sampling onto XAD-2 sorbent, specifically treated for aldehyde adsorption, followed by GC/FID analysis at SAL, Manchester.

Siloxanes: after sorbing onto XAD sorbent tubes, these were determined by GC/MS at SAL, Manchester.

Speciated Organosulfur Compounds: these were quantified by GC/MS at CERAM Research, Stoke-on-Trent.

Carbon Monoxide: this was determined on site using a portable instrument with an electrochemical cell detection

system. Additionally, an extracted sample was introduced to a laboratory situated infrared analyser.

Moisture: this was determined in accordance with the documented CPL “in-house” procedure, AM-001:1994 Pt 3 and involves the adsorption of the moisture onto silica gel. This procedure is UKAS accredited for landfill gas.

Releases to Atmosphere

Tables 9 and 10 summarise the methods for sampling and analysis of the supply gas.

Products of Combustion: A sample of stack gas was extracted via a heated sample line, fitted with an in-duct filter, and transferred to a mobile laboratory for analysis, by UKAS accredited method ML 001, utilising the following continuous analysers:

- Infrared for oxides of sulfur;
- Chemiluminescent for oxides of nitrogen;
- Infrared for carbon monoxide (with confirmatory electrochemical cell analyser for certain sites);
- Paramagnetic for oxygen; and
- Flame ionisation detector (FID) for Total VOCs

The calibration of the analysers was checked before during and after the period of testing using certified gas standards, and found to be within the operational tolerance of $\pm 4\%$.

Hydrogen Chloride: Sampling and analysis is based on USEPA method 26. A sample of the stack gases was withdrawn and bubbled through dilute sulfuric acid (1% w/v) a drying train, pump, gas meter and rotameter. The sulfuric acid sample solution was sealed and analysed at

CPL Laboratories by a UKAS accredited method utilising ion chromatography.

PCDDs and PCDFs: Polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) were sampled in accordance with the UKAS accredited in house method AM-020:1997 (Part 1d), which is based upon CEN 1948-1:1997.

Gas velocities and temperatures were measured across the two available axis to enable the determination of isokinetic flow rates. The level of accuracy for the sampling of dioxins carried out to AM-020:1997 (Part 1d) is considered to be $\pm 10\%$ (re: ISO 9096:1992).

A sample of the stack gas was removed and passed through a glass silica probe and nozzle, through a glass fibre filter paper contained in a heated oven compartment at 120°C. The filtered hot gas stream was then passed through a condenser containing a spiked XAD-2 resin trap then into an impinger train. The resin trap is spiked with an isotopic marker containing dioxins and furans, which enables a surrogate standard recovery experiment to be performed.

The condenser and resin trap were cooled below $16^{\circ}\text{C} \pm 4^{\circ}\text{C}$ with re-circulated cooled water to maintain the absorption efficiency of the XAD-2 resin trap. The impinger train is seated in a refrigerated water bath to cool the gas stream and condense out less volatile gases and vapour. The first two impingers contain de-ionised water. The third impinger is left empty to condense out any excess moisture in the gas stream. The fourth impinger contained anhydrous silica gel, to dry the gas stream, before passing through the pump and dry gas meter, which measures the volume of gas sampled. Prior to sampling, a control blank was taken. Upon completion of dioxin sampling, the filter, resin trap and solvent washings were sealed and refrigerated.

Samples, including the control blank, were submitted to an external UKAS accredited laboratory (SAL) for subsequent analysis of PCDDs and PCDFs. Analysis for dioxins and furans were undertaken on a composite sample consisting of the particulate filter, spiked resin trap and the equipment washings (toluene and acetone). High resolution gas GC/MS was employed to accurately identify and quantify the PCDD and PCDF isomers.

Polycyclic Aromatic Hydrocarbons (PAHs): The PAH sample was collected, and analysed, as part of the PCDDs and PCDFs sample, with the resin trap being spiked with PAH standard.

Hydrogen Fluoride: Sampling and analysis is based on a CERAM method. A sample of the stack gases was withdrawn and bubbled through dilute sodium hydroxide (0.5% w/v), drying train, pump, gas meter and rotameter.

The sodium hydroxide solution was sealed and analysed, at CPL Laboratories by a UKAS accredited method utilising ion chromatography.

Particulate Measurements: The particulate testing was in accordance with the CPL in-house procedure AM020:1994 Part 1, which is based upon BS3405:1983 and BS6069:1992. Gas velocities and temperatures were measured across two mutually perpendicular axes at the sampling plane to determine the relevant isokinetic flow rates and collect data required by BS3405:1983.

The silica probe was inserted into the gas flow and set to withdraw gas at the relevant isokinetic rate from each of the sampling positions.

Gases were drawn through a sharp edged nozzle and silica probe to a filter assembly containing a pre-weighed glass fibre filter. The filter assembly was maintained at a temperature of 120°C in a heated compartment.

The filtered gas was then cooled and passed through silica gel drying train, via a pump exhausting to a dry gas meter and rotameter (for a visual indication of sampling flow rate). The same filter was retained for the duration of each test i.e. cumulative sampling. After drying and conditioning, the filter was re-weighed and the mass of the collected particulate matter derived.

Oxygen: In addition to determinations made by the mobile Laboratory, stack emissions were also analysed for oxygen using portable paramagnetic or infra-red gas analysers.

Carbon Dioxide: Sampling and analysis was carried out in accordance with CPL "in-house" procedure, AM-001:1994 Pt 4 and involves the use of thermal conductivity gas chromatography (GC/TCD). This procedure is UKAS accredited for landfill gas determinations.

Moisture: A measured volume of stack gas was withdrawn through a condensing coil to a pre-weighed gas cooler and silica gel by in-house method AM-020 1994 (Part 3). After sampling, the cooler and silica gel were re-weighed and the moisture content calculated.

Speciated Volatile Organic Compounds (VOCs): The content of the gas was established via two ex-situ techniques. Firstly, low boiling range hydrocarbon compounds were analysed by packed column gas chromatography using flame ionisation detection (GC/FID) following sampling into Gresham tubes.

Secondly, the relatively less volatile compounds were identified by:

TABLE 9
SUMMARY OF SAMPLING PROCEDURES FOR EXHAUST GASES

Determinand	Sampling method	Procedure No.	UKAS Ref No.
SO ₂	IR continuous analyser	ML 001	1618
NO _x	Chemiluminescent continuous analyser	ML 001	1618
CO	IR continuous analyser, and electrochemical cell	ML 001	1618
HCl	Based on EPA 26	AM 2002	1618
PCDDs & PCDFs	CEN 1948	AM 020	1618
PAHs	CEN 1948	Not applicable	Not applicable
HF	Based on CERAM Method	AM 2001	1618
Particulate matter	BS 3405 gravimetric	AM 020	1618
O ₂	Paramagnetic and/or IR analyser	AM 020	1618
CO ₂	Gresham tubes	AM2011	1618
H ₂ O	Silica gel	AM 020	1618
VOC total	FID continuous analyser	ML 001	1618
VOC speciated	Mixed bed sorbent and Gresham tubes	AM2010/2011	Not applicable
VOC aldehydes	Treated XAD-2 sorbent tubes	Not applicable	Not applicable
Organosilicon	XAD Sorbent tubes	Not applicable	Not applicable
Organosulfur	Tedlar bag	Not applicable	Not applicable
H ₂ S	Tedlar bag	Not applicable	Not applicable

TABLE 10
SUMMARY OF ANALYTICAL PROCEDURES FOR EXHAUST GASES

Determinand	Analytical method	Analysis house	UKAS Ref No.	Sample location	Archive period
PCDDs & PCDFs	High resolution GC/MS	SAL	1549	SAL	12m
PAH	GC/MS	SAL	1549	SAL	12m
HF	IC	CPL	1618	CPL	12m
HCl	IC	CPL	1618	CPL	12m
H ₂ O	Gravimetric	CPL	1618	Not applicable	Not applicable
Particulate matter	Gravimetric	CPL	1618	CPL	12m
CO ₂	GC/TCD	CPL	Not applicable	Not applicable	Not applicable
VOC speciated	GC/MS	Harwell Sci.	Not applicable	Not applicable	Not applicable
VOC aldehydes	GC/FID	SAL	Not applicable	Not applicable	Not applicable
Organosilicon	GC/MS	SAL	Not applicable	Not applicable	Not applicable
Organosulfur	GC/MS	CERAM	0013	Not applicable	Not applicable
H ₂ S	GC/FPD	CERAM	0013	Not applicable	Not applicable

- sampling onto “mixed-bed” sorbent material (including Carboxen 1000), prior to analysis by gas chromatography with mass spectrometry (GC/MS). The sub-contract laboratory, Harwell Scientifics, analysed for up to 50 specific LQM nominated compounds considered to be potentially present in landfill gas.
- Sampling onto XAD-2 sorbent, specifically treated for aldehyde adsorption, followed by GC/FID analysis at SAL, Manchester.

Organosilicon species: after sorbing onto XAD sorbent tubes, these were determined by GC/MS at SAL, Manchester.

Speciated Organosulfur Compounds: Samples were collected into Teflon sample bags and quantified at a sub-contract laboratory (CERAM Research, Stoke-on-Trent).

Hydrogen Sulfide: Samples were collected into Teflon sample bags and quantified at a sub-contract laboratory (CERAM Research, Stoke-on-Trent).

SAMPLING PROGRAMME

Eight gas engines in the UK were sampled from six landfill sites in the period June – October 2001. The sites were chosen on the basis of a range of gas aggressiveness and to cover the three main types of nominal 1MW gas engines employed currently in the UK, as well as two larger units, including a dual fuel gas engine. The gas engines are

referred to by a reference code (E1 – E8). The data reported here represent the raw supply gas data and the emissions data at standard conditions of 273K, 101.3kPa, dry gas at 5% oxygen v/v.

While the bulk of the analyses are reported here for completeness, some of the analytes have not been considered in any great detail as yet (e.g. siloxanes, speciated VOCs) and so are not considered here further. The analysis of the data is ongoing. Further data analysis will be performed before publication of the final report on the project, so any conclusions are preliminary.

GAS SUPPLY QUALITY

The gas supply quality for the eight generating sets tested is shown in Table 11. Engines E1 and E6 are on the same landfill, and engines E2 and E5 are on another landfill site.

Bulk gases

The methane content of the supply gases ranges from 43.8% to 58.9%, with corresponding carbon dioxide ranging from 32.9% to 39.7%. The amount of entrained air introduced by the gas collection systems is relatively low, ranging from 0.18% to 3.09% oxygen, with the highest oxygen in the supply gas corresponding to the lowest methane quality, indicating that the gas quality is strongly dependent on controlling the air ingress to the gas supply. Moisture contents are also generally low (1.3% to 2.8%).

Acidic trace gases

Total sulfur in the landfill gas range over an order of magnitude, from 31 – 430 mg/Nm³ S. The bulk of this is as hydrogen sulfide (21 – 400 mg/Nm³). The site on which engines E1 and E6 were situated showed the highest sulfur in the supply gas, but this was not wholly reflected by the sulfur emissions.

Total chlorine and fluorine content can be considered for two groups of sites. Five of the landfills show halogen concentrations characteristic of mixed municipal/industrial or municipal only landfills (14.7 – 77.9 mg/Nm³ Cl (as HCl), 5.6 – 14.9 mg/Nm³ F (as HF), with one co-disposal site which has accepted halogenated organic wastes showing 557 mg/Nm³ Cl (as HCl) and 20.3 mg/Nm³ F (as HF). These higher levels of halogens in the supply gas are reflected in the emissions data below, and this is the only site investigated where these compounds exceed Ta-Luft or OAPC standards for Cl or F.

ENGINE EMISSIONS

The site data are summarised in Table 12 below. They are discussed both for the individual engine suite, since certain engine specific or site specific factors may influence the emissions, and also collectively by component or group of components in the exhaust gas.

Engine specific observations

Observations include the site aggressiveness index, where $I = ([\text{total Cl/mg/Nm}^3] + 2[\text{total F/mg/Nm}^3]) \times 100 / [\text{CH}_4/\%]$ for the supply gas quality.

Engines E1 and E6: The landfill gas at this site has a gas aggressiveness index of 202 (an aggressive landfill gas site), with the additional impact of sulfur and organosilicon compounds in the supply gas. Generating set E1 was recently overhauled and set E6 was requiring complete overhaul when sampling was carried out.

Very little difference was seen between the two engines in terms of NO_x (set via the engine management system), PCDDs/PCDFs, HF or total VOCs/methane slippage. CO emissions were high from both gas engines, 1800 mg/Nm³ on set E1, but 2600 mg/Nm³ on set E6. There was also a significant difference between the two engines in SO_x, at 45mg/Nm³ on set E1, and 120mg/Nm³ on set E6. The latter figure is partially attributed to lubricating oil passing the worn piston rings, rather than the effect of gas quality.

Engine E2: The landfill gas at this site has a gas aggressiveness index of 186, due mainly to chloride compounds in the inlet gas, although this is not obvious from either the HCl or PCDD/PCDF emissions. Both NO_x (760 – 820 mg/Nm³) and CO (1400 – >1500 mg/Nm³) emissions were high.

Measurements were taken at this site both just after the turbocharger and using the stainless steel extension sleeve. The sampling sleeve achieved very stable laminar flow across the sampling plane. This suggests that pollutants in the gas stream should be well mixed in a relatively homogeneous manner, and the sleeve is therefore an ideal solution for measuring PCDDs/PCDFs, PAHs and particulates, since isokinetic sampling conditions were successfully achieved.

Bulk gas concentration measurements are, however, very close to the results obtained at the turbocharger outlet, and not a large difference was seen between results for VOCs and other trace constituents either. It is likely that the sampling sleeve will therefore only be needed for situations where isokinetic sampling is required, and not for routine emissions sampling.

Engine E3: The landfill gas at this site has a gas aggressiveness index of 44 (a mildly aggressive landfill gas site). As with most gas engines sampled, there was methane slippage and incomplete combustion of the supply gas: VOCs (2350mg/Nm³) and CO (1520mg/Nm³) were high. Since chlorine was low in the supply gas, PCDDs and PCDFs were close to the detection limit.

TABLE 11.
BULK AND TRACE GASES IN THE SUPPLY GAS AT THE GAS GENERATING SETS STUDIED

Engine reference no:	Gas Generating Sets								
	E1	E2	E3	E4	E5	E6	E7	E8	
Engine type	SI	SI	SI	SI	Dual fuel	SI	SI	SI	
Analyte	Units								
Methane	%	43.8	56.8	58.9	52.1	56.8	43.8	56.6	45.5
CO₂	%	32.9	39.7	36.9	36.4	39.7	32.9	37.1	33.8
Oxygen	%	3.09	0.18	0.54	0.65	0.18	3.09	0.82	2.00
Hydrogen	%	0.03	0.07	0.02	0.08	0.07	0.03	<0.01	<0.01
Moisture	%	1.69	2.34	1.26	4.11	2.34	1.69	1.64	2.82
Total S	mg/Nm ³	166.5	30.8	33.0	39.8	30.8	166.5	184.1	430.5
Total F	mg/Nm ³	14.9	<13.9	5.6	20.3	<13.9	14.9	<13.9	<13.9
Total Cl	mg/Nm ³	58.6	77.9	14.7	557	77.9	58.6	76.9	23.9
Organo S	mg/Nm ³	<22.9	<27.0	<8.0	<119.9	<27.0	<22.9	<10.1	<11.3
H₂S	mg/Nm ³	110.8	65.3	21.4	93.6	65.3	110.8	144.2	400.0
Organo Si	mg/Nm ³	10.7	5.3	<6.7	28.6	5.3	10.7	148	5.5
CO	mg/Nm ³	146	84	22	139	84	146	28	30
Total NMVOC	mg/Nm ³ ¹	983	1221	565	nd	1221	983	181	nd
	mg/Nm ³ ²	<626	633	426	1440	633	<626	<198	<120

Notes SI – internal combustion spark ignition engine. Dual fuel – diesel fuel pilot ignition engine. nd – not determined.

Gas quality is as received at the generating set. Concentration results are expressed in mg/Nm³ (273 K, 101.3 kPa, moist gas). Percentages expressed by volume (% v/v) on dry gas basis.

NMVOCs are (1) by FID from Gresham tube samples, and (2) sum of individual NMVOC species identified by GC/MS and GC/FID.

TABLE 12
BULK AND TRACE GASES IN THE EXHAUST GASES AT THE GAS GENERATING SETS STUDIED

Engine	Gas Generating Sets								Standards		
	Reference no:	E1	E2	E3	E4	E5	E6	E7	E8	TA-LUFT	OAPC
Engine type	SI	SI	SI	SI	Dual fuel	SI	SI	SI	1986	1988	
<i>Analyte</i>											
SO _x	45	74, 88	18	60	540	120	110	160	500	250	
NO _x	360	820, 760	370	1260	1500	500	490	720	500	400	
CO (continuous IR)	>1500	>1500, 1400	1520	508	>1900	>1500	1050	1400	650	650	
CO (electrochemical cell)	1800	nd	nd	nd	nd	2600	nd	nd			
HCl	9.5	5.7	0.9	584	1.7	1.6	0.2*	3.0	30	30	
HF	3.1	<1	<0.4	45	<1.3	3.2	3.8*	6.2	5	5	
PCDDs/PCDFs	0.0011	0.0046	0.0027	0.013	nd	0.0012	0.0026	0.0009	-	-	
PAHs	0.026	0.023	0.01	0.003	nd	0.0018	*	0.0049	-	-	
Particulates	6.7, 4.2	4.4, 3.5	2.6, 2.1	7.5, 6.3	33, 18	4.1, 2.9	2.0, 1.7	4.9, 7.2	50 / 150	50	
CO ₂	15.4	17.6	18.0	17.0	14.3	15.8	13.1	17.6	-	-	
Total unburnt hydrocarbons	1300	1100, 1200	2350	590	5260	1200	530	1700	-	-	
NMVOC	20	23	71	9	66	19	48, 39*	1	20 - 150	20 - 150	
Methane	1700	680	2450	690	5470	1840	2740, 2056	840, 2490			
Organo S	<7	<7	<7	<9	<7	<7	<7	<7			
H ₂ S	<3	<3	<3	<3	<3	<3	<3	<3			
Organo Si	<1.0	<0.6	<0.1	<0.5	<1.0	<1.2	<0.5*	<0.6			

Notes

All measurements are in mg/Nm³ except for PCDD/PCDF which are in ng/Nm³ TEQ and CO₂ which are in %.

All measurements are corrected to standard temperature and pressure conditions, dry gas, 273K, 101.3 kPa, 5% oxygen.

SI – internal combustion spark ignition engine. Dual fuel – diesel fuel pilot ignition engine. nd – not determined. * to be confirmed.

Total unburnt hydrocarbons are reported as mg (carbon)/Nm³. NMVOCs or methane are the sum of individual VOCs or methane as mg/Nm³.

Engine E4: The landfill gas at this site has a gas aggressiveness index of over 1100 (a very aggressive landfill gas site), due mainly to a very high chloride loading (557mg/Nm³ total Cl). Sulfur is also high, at 94mg/Nm³ hydrogen sulphide.

Emissions of NO_x were high (1260mg/Nm³) but CO was low (508mg/Nm³). This site is also the only site to exceed TA-Luft limits for halogen emissions (584mg/Nm³ HCl and 45mg/m³ HF compared to thresholds of 30mg/Nm³ HCl and 5mg/Nm³ HF under TA Luft, 1986).

Whilst all other sites have PCDDs/PCDFs close to the detection limit, the presence of high concentrations of chlorine and fluorine compounds in the supply gas do appear to promote formation of more PCDDs/PCDFs than at landfills with more typical Cl/F loadings.

Dual-fuel engine E5: The landfill gas at this site has a gas aggressiveness index of 186, due mainly to chloride compounds. This diesel fuel pilot ignition engine, situated at the same site as engine E2, is of a different design to the other spark ignition engines tested. NO_x (1500mg/Nm³), CO (>1900mg/Nm³) and SO_x (540mg/Nm³) emissions are all high, as are total unburnt hydrocarbons (5260mg/Nm³). Some of the SO_x will be derived from the diesel pilot fuel, and the relatively high lubricating oil consumption.

HCl emissions do not reflect the inlet gas as much as emissions from set E2. The sample port arrangements did not allow measurement of PCDD/PCDF/PAH/particulates.

Engine E7: The landfill gas at this site has a gas aggressiveness index of 185 due to chlorine. High CO (1050mg/Nm³) and methane slippage (2000 - >2700 mg/Nm³) were recorded, but otherwise low emissions.

Engine E8: An aggressive gas site in respect of chlorine and fluorine (a gas aggressiveness index of 185), but also with the highest sulfur levels in the inlet gas of all sites tested gave the highest SO_x emissions (160mg/Nm³) with the exception of the dual-fuel engine. PCDD/PCDF emissions were therefore low and close to detection limits.

Emissions Data - Preliminary Interpretations

NO_x, CO and unburnt hydrocarbons: Carbon monoxide is the product of incomplete combustion, and reduces as the combustion efficiency increases. Conversely, NO_x increases as the thermal efficiency of the engine increases. This is because the peak combustion temperature increases as combustion efficiency increases, and formation of NO_x is related directly to the peak combustion temperature. Destruction of VOCs is better achieved with higher combustion efficiencies.

This project has found that CO emissions are commonly

far higher than TA-Luft and OAPC standards when nominal 1MW gas engines are tuned to approach the TA-Luft or OAPC NO_x limit. Variation of engine control settings, such as fuel admission timing, turbocharger air flow and air/fuel ratio, can alter emissions somewhat. A short test to increase combustion temperature increased NO_x, reduced CO and reduced VOCs at site E3. These adjustments are, however, refinements to the principal engine design whereby bore, stroke, compression ratio and speed have a significant influence on emissions.

CO is considered to be of less concern than NO_x in relation to global warming and it is understood that the TA Luft limit for CO may be raised in March 2002. CO, which will subsequently oxidise to CO₂, is typically 1% of total CO+CO₂ emissions.

Between 97% to 99% of VOCs in emissions have been shown to be unburnt methane. Further work is being carried out on this aspect of engine emissions, but destruction efficiencies of ≥99% are found for methane and all individual NMVOC species above detection limits.

Acid forming compounds

High concentrations of potentially acid forming functional groups containing Cl, F and S in the supply gas should theoretically be reflected in higher HCl, HF and SO_x exhaust emissions. Unplanned burning of lubricating oil will also be a source of SO_x in the exhaust gas, as indeed could be burning of pilot diesel fuel in dual fuel engines.

The mass balance for Cl, F and S compounds is reasonably consistent, suggesting that these elements tend to remain in the gaseous phase, forming simple acid gases HF, HCl and SO₂ during the combustion process, and which are found in the exhaust emissions. The presence of Cl and F in the supply gas is also linked to PCDD/PCDF formation (see below). High concentrations of chlorine and fluorine in the inlet gas does appear to result in higher emissions of PCDDs/PCDFs, although the relationship is only clear with very high chlorine/fluorine loadings in the supply gas.

The bulk of the sulfur emissions are derived from hydrogen sulfide in the supply gas. The dual-fuel engine has a higher lubricating oil consumption compared to others and this, combined with the diesel pilot fuel, may be partially reflected in the high SO_x emissions (540mg/Nm³) compared to the relatively low SO_x in the supply gas. A similar situation was observed on engine E6, where piston overhaul was imminent.

PCDDs and PCDFs:

PCDDs and PCDFs are products of incomplete combustion of large halogenated organic compounds (such as chlorinated solvents, CFCs, etc) and the subsequent reformation into PCDDs/PCDFs post combustion. They

are likely to increase with increasing measured total Cl and F in the supply gas, but this relationship only becomes apparent on sites with a very high gas aggressiveness. Many site data for PCDD/PCDF emissions are effectively at the limit of detection of the instrumentation used.

The highest recorded value was 0.013ng/Nm³ PCDD/PCDF measured at site E4. 0.005 ng/Nm³ is a more typical maximum value from a household waste site. The limit of detection for the method employed is typically between 0.001 – 0.003ng/Nm³TEQ.

Existing UK limits for PCDDs/PCDFs in emissions from furnaces burning refuse-derived fuel (HMSO, 1995) are from 0.1 – 0.5 ng/Nm³. The European Commission (2001) propose a threshold of 0.1 ng/Nm³ for gas engines burning biogas (landfill gas is specifically excluded from the draft 2 definition of biogas) PCDDs and PCDFs present in the exhaust stack of landfill gas engines appear to be well below the threshold concentrations for analogous plant emissions, even at very aggressive gas sites.

CONCLUSIONS

High temperature combustion, by utilisation of landfill gas, typically destroys ≥99% of the NMVOCs in landfill gas, as well as methane. This potentially reduces the local and global environmental impacts. However, some combustion products are formed which have local environmental impacts. The findings reported in this paper are preliminary, and a final report will be issued in mid 2002. These are the key indicators to date.

High concentrations of potentially acid forming functional groups containing Cl, F and S in the supply gas tend to reflect higher HCl, HF and SO_x exhaust emissions. Unplanned burning of lubricating oil may also be a source of SO_x in the exhaust gas, as may be burning of pilot diesel fuel in dual fuel engines. However, only the highest chlorine loading found in this study exceeded the TA-Luft standards for HCl emissions, and only the dual-fuel gas engine tested exceeded the SO_x limit.

PCDDs and PCDFs are products of incomplete combustion of complex halocarbons and may form post combustion. Their presence is believed to be at least in part proportional to the measured total Cl and F in the supply gas. However, all measurements, even at the highest chlorine loadings in the supply gas, are below current UK standards for refuse derived fuel plant, and proposed EU standards for engines running on biogas (which in the latest draft specifically excludes landfill gas). Many measurements were at or close to the detection limit of the technique. It is expected that measurement of PCDDs/PCDFs, which is an expensive technique, will not therefore be a routine requirement, even at aggressive gas sites, in any proposed standards for landfill gas engines. Some countries regulate

on the gas quality of the supply gas, and such an approach seems valid for screening.

Valuable information has also been collected regarding typical engine performance tuning, which is currently based on attaining optimum thermal performance – without the need to meet any specific emission limits for NO_x, CO, or unburnt hydrocarbons. Further work is being carried out on this aspect of the research, and also on the detailed analysis of the trace gas component inventory and the impact of organo-silicon compounds.

Working alongside the regulator, we expect this research will help in the development of appropriate gas engine emission monitoring protocols and standards in future UK technical guidance.

REFERENCES

- Couth, B., 2001. "To investigate the Environmental Effectiveness of Landfill Gas Management in England and Wales and to assess how it should be improved". MSc Dissertation, De Montfort University, Leicester. 2001.
- Environment Agency, 1999. Interim Internal Technical Guidance for Best Practice of Landfill Gas. Consultation Draft. Document No. LFG2, Version 2.0, March 1999.
- EPL, 1997. Federal Law relating to the Protection of the Environment of 7 October 1983, latest amendments passed 21 December 1995 (as of 10 June 1997). 814.01.
- European Commission, 2001. "Biological Treatment of Biodegradable Waste". Working Paper Draft 2, Directorate A.2, Sustainable Resources.
- HMSO, 1995. Processes subject to Integrated Pollution Control. IPC S2 1.05. Combustion Processes: Combustion of Fuel Manufactured from or Comprised of Solid Waste in Appliances in 3MW_{th} and Over. London: HMSO September, 1995.
- OAPC, 1998. "Ordinance on Air Pollution Control of 16 December 1985" as at 3 February 1998.
- Scott, P., and Haigh, C., 1999. "Emissions from Landfill Gas Energy Recovery Plant - Monitoring Protocols". Entec UK Ltd, Environment Agency R&D Technical Report P248 for contract CWM 181/97.
- TA-Luft, 1986. "First General Administrative Regulation Pertaining the Federal Immission Control Act" (Technical Instructions on Air Quality Control - TA-Luft) as of 27 February 1986 (GMBI. P.95, 202).