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Odours & Voc Impact Evaluation Of Three Gategories Of Landfills

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Emissions from municipal solid waste landfills (MSWL) have the potential to cause olfactive annoyance and health impacts among the surrounding populations. The objective of this study was to examine the impacts on the air quality of MSWL in Quebec in terms of odours and potentially toxic compounds related to landfill gas emissions. The project was performed on six MSWL through an assessment of odours and potentially toxic emissions and dispersion modelling at each landfill site to estimate the human exposure to potentially toxic compounds and odours. The emissions of selected pollutants, such as volatil organic compounds (VOCs), terpens and sulfur compounds, were estimated by using the landfill gas emission LandGEM 2.01 software and by a methodology based on field measurements. The exposure to pollutants was estimated by emission assessment using the field measurements methodology combined with two atmospheric dispersion models: the Gaussian model of ISCST3 software and the Gifford-Gaussian model of TROPOS software which is specifically designed for odour dispersion. The meteorological conditions under which the dispersion models were applied referred to the representative scenario in southern Quebec. The assessed air pollutant concentrations for potentially toxic and odorous compounds were significantly below the Quebec exposure health criteria.

1. Introduction

Methane in municipal solid waste landfills (MSWL) is produced through anaerobic microbial degradation of organic matter. When the conditions of methanisation are stable in wastes, landfill gas is mainly made up of methane (CH_4) and carbon dioxide (CO_2) in about equal proportions. The stable production of methane, interesting for valorisation, can last several years. Biogas contains others gases in traces, primarily sulfur compounds and volatile organic compounds (VOCs).

The methane and the carbon dioxide are greenhouse gases like some trace gases in landfill gas, as for example the chlorinated and fluorinated organic compounds. Landfill gas has other effects on the environment: in addition to the explosive potential of methane, important risks and harmful effects to the environment and public health exist because of the presence of hydrogen sulfide (H_2S) and of potentially toxic VOCs (benzene, vinyl chloride, dichloromethane, chloroform, toluene, dichlorobenzene, etc) as well as compounds responsible for odours (VOCs, sulfur compounds, etc).

The overall objective of this study was to examine the impacts on the air quality of

MSWL in Quebec in terms of odours and potentially toxics compounds related to landfill gas emissions.

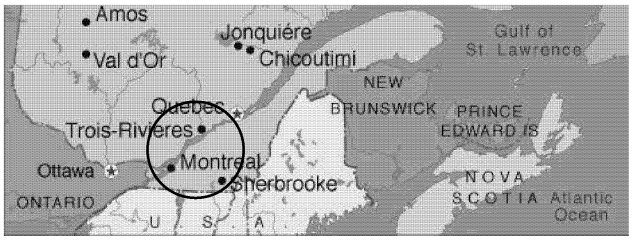
2. Methodology

2.1 Experimental procedure overview

In order to evaluate the impacts on air quality of six landfills representative of the Quebec landfills, six stages were proposed:

1. Scientific planning,
2. Experimental measurements,
3. Evaluation of the impacts,
4. Possibility of identifying some(s) tracer(s),
5. Review of the regulation applicable to landfills and their impacts on air quality,
6. Conclusions and recommendations on the methods to monitor and evaluate the impacts on air quality.

The six MSWL represented the three categories defined by the Québec Ministry of Environment (MDDEP) according to the landfilling capacity. The three categories were considered because the small sites without active collection of landfill gas represent a considerable share of the total emissions associated with landfilling activities in Quebec.



To compare the landfills, each site was represented according to a similar theoretical configuration. For each site, the emissions were characterised on two types of sources: the collected caped surfaces and the landfill gas collection system. Other sources of contaminants and odour potentially present on a landfill (fresh waste, system of lixiviates management, etc) were not taken into account in this study.

The planning of field tests comprised four stages: landfills selection, preliminary characterisation, planning of experimental measurements, and calculation by modelling of the generated landfill gas flows.

The characterisation of landfill gas was performed by sampling in the landfill gas collection system. Then, the surface sampling comprised cartography of the methane surface concentrations and establishment of the correlation between methane surface concentration and methane surface flux (Fécil et al., 2003). The methane surface concentration cartography was performed according to the recommendations of the instantaneous surface monitoring (ISM) method described in the Californian rule 1150.1. The methane surface flux was measured with a dynamic flux chamber operated according to the recommendations of the US EPA (Klenbusch, 1986).

The concentrations measurements in pure landfill gas and the surface emissions were then compared in order to lay down the applicable rule of proportionality. Surface emission rates of the various compounds measured in pure landfill gas were then estimated on the whole of the site using the proportionality rule and the correlation between the methane surface concentration and the methane surface flux.

The landfill gas flow generated on each site was calculated first with the landfill gas emission LandGEM 2.01 software, which includes the landfill gas composition data from AP-42. The emission rates of the studied compounds were thus evaluated on each site by two approaches: the generation modeling with LandGEM and the field measurements methodology, which used the landfill gas characterisation described above and the actual landfill gas flow measured on site. The evaluation of actual landfill gas flow rate used gas recovery data when available at the site. Otherwise, landfill gas flow rate was evaluated with field measurements of velocity in the landfill gas collection system. A comparison of the two approaches was thus carried out on the six sites.

The study of the impacts on the air quality proceeded in four stages: the compilation and the interpretation of the emission data, the choice of the meteorological data, the modeling of atmospheric dispersion, and the interpretation of the dispersion modeling results. The interpretation of the results comprised a preliminary toxicological analysis.

The possibility of identifying one or several integrating compound(s) was then evaluated using the whole of the results. The strategy adopted at this stage was dictated by the results of the emissions characterisation and by the results interpretation on the whole of the sites.

The examination of the relevant regulations throughout the world was carried out in order to identify the regulations applicable to the landfill gas issues in MSWL and to their impacts on the ambient air quality.

2.2 Experimental methods

The characteristic parameters of landfill gas were identified according to their impact on air quality (potential toxicity, respect of the standards, potential olfactive nuisance). The landfill gas compounds identified like odorous are mainly terpens (limonene, careens, camphene, pinene, phellandrene, etc.), sulfur compounds (mercaptans, sulfides) and some VOCs (ethylbenzene, styrene, toluene, benzene, etc).

The odour of landfill gas was also characterised in term of odour concentration because, taking into account the non-additivity of the odour of each component in a mixture (Gostelow et al., 2003), the great variability of the odour perception thresholds reported in literature (AIHA 1989) and the inexistence of certain values of these thresholds, only a sensory analysis is able to account for the effects of interactions in the cocktail of compounds which is landfill gas. The odour concentrations cannot be deduced from the chemical composition of landfill gas and of the odour perception threshold of each compound. Then, the determination of the odour concentration requires the measurement by olfactometric analysis. Olfactometry consists in measuring the odours according to a standardized and recognized procedure, in order to characterise the odours emissions.

The 40 VOCs analysed by method TO-14A of the US EPA are potentially toxic compounds, of which some are emitted by MSWL and are to be considered because of

their potential of toxicity.

The sampling techniques implemented in the project were as follows:

? Sampling out of Tedlar bags or passivated canisters for the VOCs,
? Sampling out of Tedlar bags for the odours and the sulfur compounds,
? Measurement of the surface flux rates with a dynamic flux chamber.

The emission measurements were thus carried out at the source: samples were collected at the source and sent from the site to the laboratory, in order to analyse them in controlled and standardised conditions. The odours were measured within 30 hours of collection to minimise sample losses, degradation or alteration.

Ambient air sampling is generally not recommended because of the frequent variations of concentrations (Myles and Mason, 1991). Field measurements of odour in ambient air have a weak resolution, mainly because of the lack of precise knowledge of the micrometeorological conditions. Moreover, sensory odour measurements lack the sensitivity necessary to measure low odour concentrations at downwind locations. Thus, it is difficult to carry out sampling or measurements of odours in ambient air. The odours perceived in ambient air are due to the odour emissions from sources and atmospheric dispersion. The measurement of the odour concentration at the source is a method making it possible to evaluate the odour concentration in ambient air following a simulation of atmospheric dispersion.

These considerations apply to the odours but also to the compounds measured by chemical analyses.

The measurement of surface flux rates in dynamic flux chamber is very powerful for the landfills and easier to implement than the wind tunnels. The method of the dynamic flux chamber is a method of non intrusive and non destructive direct measurement. Its principal advantages compared to the other measurements methods are the simplicity of implementation, the flexibility and the precision. Dynamic flux chamber is easy to implement, but wind tunnels allow studying the variation in emission with sweep velocity.

The analytical methods used in this project are adapted to MSWL emissions characterisation and they are powerful and recognized techniques:

? Portable FID, GC-FID or GC-TCD for the methane and the carbon dioxide,
? GC-MS with cryogenic trap for the VOCs and the other organics like terpens (method TO-14A of the US EPA),
? GC-PFPD for the sulfur compounds,
? Olfactometry with dynamic dilution (methods ASTM E679, EN 13725, probit) for the odours.

In this project, odour measurements were performed by the two usual approaches, sensory analyses and analytical techniques. Sensory tests concern the perceived effect of the odorants mixture as detected and interpreted by the human olfactory system. Analytical methods or physicochemical analyses relate to the properties of the odorants.

The landfill gas production is governed by many and complex phenomena. Various models exist to represent the kinetics of waste biological breakdown and to thus estimate the kinetics of landfill gas production. LandGEM is a simple kinetic model of order 1 which proposes default values for the entry parameters. It is however more

powerful with values specific to the studied site. Two other tools for simulation of the generation with a simple kinetic model of order 1 were also considered: the E-Plus software and the Huitric and Soni (1997) method. The landfill gas flow emitted by each site was estimated with the assistance of these three tools and was confronted with the experimental data.

According to the regulation review on the atmospheric emissions of the MSWL, the legislator requires generally the use of the best techniques available to control the emissions. The American states have very variable regulatory requirements for the chemical follow-up at the source and in ambient air. In the European countries, the follow-up on the chemical compounds is carried out at the source. Germany and the Netherlands have the most advanced regulations based on criteria of frequency of exposure to odour concentrations.

3. Results And Discussion

Work completed made it possible to conclude on the impact associated with the landfill gas emissions from the sites according to the site size and to make recommendations on the target contaminants and the methods and approaches of characterisation and monitoring.

Site		CH4 generation LandGEM	CH4 measured	Ratio
Nº	Cat.	[106 m3]/yr	[106 m3]/yr	[%]
#1	A	32,90	36,71	112%
#2	A-B	21,67	14,86	69%
#3	B	7,89	10,76	136%
#4	B	4,64	0,18	4%
#6	B-C	2,07	3,28	159%
#7	C	0,47	0,37	78%

AP-42 default values
K = 0.04 year-1
L0 = 100 m3/Mg
K: methane generation rate constant (year-1)
L0: methane generation potential (m3 of CH4/Mg of refuse)

For the studied contaminants, H_2S like the potential carcinogens, the results of the study showed that the impact on the air quality is in conformity with the criteria or standards of air quality considered, these criteria being recorded in official sources, that is to say the US EPA and the MDDEP. According to the results, the odours represent the principal impact of the sites on the air quality.

For the contaminants and the odorous compounds, the field values and the emission factors of the AP-42 did not identify the same compounds as priority odorants. In the case of the carcinogens, each site presented different dominating compounds. The dominating odorous compound would be H_2S on all the sites according to odour threshold values retained for calculations. However, the analytical measurement of odorants underestimated of two orders of magnitude the odour concentration compared to olfactometric measurements.

According to the experimental results, the composition of landfill gas varied from one site to another and differed from that proposed in the AP-42. Thus, the measurement of the landfill gas composition on each site is recommended for the emissions characterisation of contaminants and odorants rather than the use of the AP-42.

		(1)	(2)	(3) & (4)					
Composé (#)	CAS (#)	Coef. $(\mu g/m^3)^{-1}$	AP-42 LES-X	Mesures biogaz réseau LES-1	LES-2	LES-3	LES-4	LES-6	LES-7
Chlorure de vinyle	75-01-4	8,80E-06	21,0%	35,7%	0,2%	6,7%	20,1%	0,2%	3,2%
1,1-Dichloroéthylène	75-35-4	5,00E-05	5,0%	3,3%	3,2%	5,9%	0,7%	3,0%	7,0%
Dichlorométhane	75-09-2	5,00E-07	3,2%	0,5%	0,0%	0,1%	0,4%	0,0%	12,0%
Chloroforme	67-66-3	2,30E-05	0,4%	-	3,8%	3,8%	-	2,1%	1,1%
1,2-Dichloroéthane	107-06-2	2,60E-05	5,5%	0,6%	0,9%	8,7%	-	1,9%	0,0%
Benzène	71-43-2	7,80E-06	6,1%	10,8%	0,1%	9,7%	77,0%	0,4%	26,5%
Tétrachlorure de carbone	56-23-5	1,50E-05	0,0%	-	2,3%	-	-	2,9%	-
Trichloroéthène	79-01-6	4,35E-07	0,8%	0,4%	0,0%	0,1%	0,2%	0,0%	1,0%
1,1,2-Trichloroéthane	79-00-5	1,67E-05	1,2%	2,5%	-	-	-	2,6%	17,7%
1,2-Dibromoéthane	106-93-4	5,00E-04	0,5%	35,3%	-	55,7%	-	73,2%	0,0%
1,1,2,2-Tétrachloroéthane	79-34-5	5,80E-05	56,3%	8,4%	87,5%	6,8%	0,0%	9,2%	13,7%
Hexachlorobutadiène	87-68-3	2,20E-05	-	3%	2%	3%	2%	4%	18%

(1): IRIS database. Carcinogen risk from inhalation exposure (continuous lifetime exposure from birth).

(2): AP-42 emission factors for LFG constituents (LFG at 55% CH4)

(3): 45% preponderant carcinogen in total risk index

(4): 7% carcinogen accounting for more than 5% in total risk index

The results showed that the odour emissions should be established with the odour concentration determined by dynamic dilution olfactometry and not with the odorous compounds, whether the odorants concentrations are determined by the values of the AP-42 or measured in situ.

		A	A-B	B	B-C	C
	AP-42	#1	#2	#3	#6	#7
H2S	79%	87%	93%	97%	89%	85%
TRS	91%	89%	95%	98%	97%	94%
Other Sulfur	8%	3%	2%	1%	0%	2%
VOC	0%	0%	0%	0%	1%	0%
Terpenes	-	8%	3%	1%	1%	4%
H2S (o.u./m3)	71 167	109 422	172 937	186 905	9 050	51 020
Odour (o.u./m3)	-	1 454 000	629 000	2 151 000	41 000	233 000

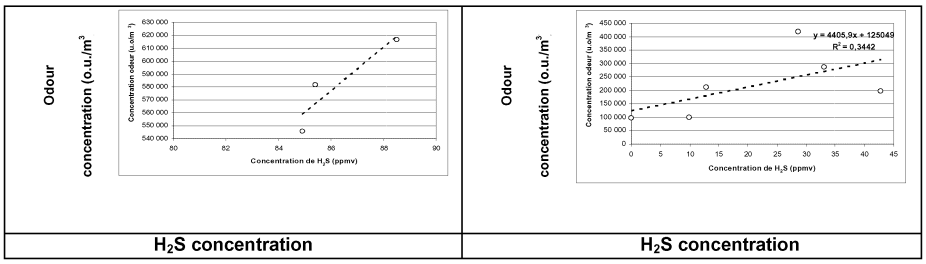
The limit of sensitivity (about 20-50 ou/m^3) and the repeatability (poor for concentrations lower than 1,000 ou/m^3) limit the applicability of olfactometry, and make it unsuitable for ambient odour assessments (Gostelow et al., 2003). Nevertheless, the sensory determination of the odour concentration provides comparable data for different emissions sources and can provide input data for dispersion models, to determine odour impact in terms of annoyance and to assess the efficiency of abatement measures (Bockreis et al., 2005).

Generally, it is difficult to characterise properly odour using analytical measurements for atmospheric pollutants. Analytical measurements characterise odours in terms of their chemical composition and the quantification of the odorants by their perception threshold. The number of odorants is very large in landfill gas as in other

environmental odours, and the odorants present were at concentrations lower than or close to detection limits. The relationship between the concentration of odorants and the perceived odour is difficult to establish, as synergistic and antagonistic effects are observed between odorants (Hallier et al., 2004). This is specially the case for complex mixtures of odorants as environmental odours. Moreover, the differences in values of odour thresholds in literature are very considerable, typically showing a range of several orders of magnitude.

Compared to olfactometry, analytical methods offer greater accuracy and repeatability and the quantified odorant components can be linked to odorant production mechanisms. However, the olfactometry is recommended because of the previously described disadvantages of analytical measurements.

In this project, the preponderant odorant was hydrogen sulphide (H_2S) according to the chemical analyses in pure landfill gas; however it was not possible to establish a correlation between odorants concentrations and odour concentration.



According to the emissions evaluation results, on the sites with an active collecting system and from average size to large, the effort must relate to the characterisation and the quantification of the surface emissions. On the sites with a passive collecting system, the effort must relate to the complete characterisation of landfill gas in the network and the quantification of the flows to the vents.

	Odour flow rate			Total area (m2)	Total land filled (106 tons)	o.u./s per ton
	Total (o.u./s)	Coll. Syst (%)	Surface (%)			
Active collection (Site 1, type A)	132,000	21%	79%	575,000	13	0,01
Passive collection (Site 7, type B)	5,800	80%	20%	44,000	0.25	0,02

The criteria of selection suggested to define a potential tracer were the olfactive properties, toxicity in terms of cancerogenicity, the facility of sampling, the facility of analysis in laboratory and interferences and background noise. On the basis of work completed, it appeared that there was no tracer compound which could represent the contaminants or the odour of the MSWL landfill gas for all the six landfill sites.

4. Conclusions

The evaluation of the impact on the air quality of the contaminants and the odours associated with the MSWL landfill gas emissions should relate to the characterisation and the quantification of the emissions at the source combined with a modeling of atmospheric dispersion.

Since the principal impact identified in this research was the odour, it is thus recommended to carry out a regular or continuous monitoring of odour emissions in term of odour concentration odour.

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