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ORIGINAL PAPER

Evaluation of risk assessment of landfill emissions and their impacts on human health

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Abstract
This study investigates emissions from landfills and the effect these emissions may have on human health. Landfill emissions contain methane, carbon dioxide, malodour compounds, and volatile organic compounds (VOCs). VOCs comprise a specific class of air pollutants that are potentially hazardous not only to the environment but also to human health. Landfill workers along with residents living in close proximity to the landfill are at risk of inhaling ambient VOCs, which potentially may cause acute or chronic illness. VOCs can have a negative effect on human health. For example, benzene, toluene, ethylbenzene, and xylenes, which together form BTEX, are individually and collectively human carcinogens, including at low concentrations. BTEX are the main compounds of VOCs that can be identified and quantified in the landfill biogas. The main objective of this paper is to estimate the concentration of BTEX and their risk assessment on human health. An emitted biogas was collected from the surface of a passive methane oxidising bio-cover (PMOB) installed in Saint Nicephore (Quebec, Canada) landfill since 2006. The sampling period was taken place from June to September where the temperature is in the range of 16 to 30 °C approximately. The biogas collected was treated in the laboratory the same day. A gas chromatograph coupled with a mass spectrometer is used to identify and quantify BTEX compounds. The concentrations of BTEX are in the range of below the limit of detection (BLD) to 3.76 mg m^{-3}. The intake rate (IR) values ranged from 0.27 to 0.39 mg/kg-day, while the estimated values of the cancer risk directly attributable to the landfill's VOCs ranged from 0.007 to 0.010.

Keywords BTEX · Landfill biogas · Human health · Risk assessment

Introduction

The term "landfill" is used to describe a unit operation for the final disposal of municipal solid waste (MSW) on land. Generally in the north of America, landfills are still one of the predominant methods for disposing and treating MSW (Staub et al. 2011). The cheapest method, in terms of exploitation and capital costs when compared to other methods (e.g. incineration, composting, etc.), is landfilling. This strategy, however, produces biogas emissions, which then rise into the atmosphere and cause air pollution. The decomposition of solid wastes may cause further environmental problems if biogas emissions are not controlled. Solid wastes contain a wide variety of hazardous substances, such as vehicle maintenance products, mercury, toxic detergents, pharmaceuticals, garden pesticides, batteries, and other industrial by-products (Slack et al. 2005). Despite ever-increasing calls for environmental protection, the dumping of solid waste, particularly in landfills, remains a prominent means of disposal and implied treatment (Hamer 2003).

Municipal solid waste is composed of 40 to 50% cellulose, 9 to 12% hemicellulose, and 10 to 15% lignin on a dry weight basis, with the cellulose plus hemicellulose (carbohydrates) accounting for about 90% of the biodegradable fraction (Barlaz et al. 1989, 1990). According to Barlaz et al. (1990), when MSW is buried in a landfill, a complex series of chemical and microbiological reactions is initiated. Biodegradation of solid waste contributes to a meaningful reduction of its volume, energy recovery in

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the form of biogas, and compost production (Liwarska-Bizukojc and Ledakowicz 2003). Landfill biogas (LFG) is produced by microbial degradation of organic matter under anaerobic conditions. Anaerobic degradation can be defined as a biological conversion process without external electron acceptors such as oxygen in aerobic processes or nitrate/sulphate in anoxic processes. Biogas is a pollutant characteristic of MSW in landfills (Aronica et al. 2009). The alternative treatment technologies for solid waste management are subject to criticism either by environmentalists (on the grounds of possible hazardous emissions, failure to eliminate pathogenic agents, or failure to immobilise heavy metals) or by landfill operators and contractors (on the basis of waste management economics). Some common volatile organic compounds (VOCs) are found in landfill biogas (LFG) composed of benzene, toluene, ethyl-benzene, and xylene isomers (known by BTEX) (Badjagbo et al. 2010; Durmusoglu et al. 2010; Yaghmaien et al. 2019). These are commonly referred to as BTEX. The BTEX compounds form an important group of VOCs because of their deleterious effect on the tropospheric chemistry and their neurotoxic, carcinogenic, and teratogenic properties (Durmusoglu et al. 2010). Based on epidemiologic studies, VOCs have been proven harmful to populations that inhale the polluted air (Nair et al. 2019). Furthermore, exposure to VOCs has been linked to a variety of negative health outcomes (Dai et al. 2017; Nair et al. 2019). The risks associated with biogas emitted by landfills have not been fully elucidated, nor have the risks to populations (workers) posed by the inhalation of hazardous air pollutants been well-characterized (Gong et al. 2017; Yaghmaien et al. 2019).

According to several researchers (Durmusoglu et al. 2010; Dai et al. 2017; Gong et al. 2017; Masih et al. 2017; Kumar et al. 2018; Zhang et al. 2018; Garg and Gupta 2019; Rafiee et al. 2019), BTEX is considered a carcinogenic substance that is known for its ability to deteriorate human health. Individually, both benzene and ethyl-benzene are carcinogens as well, affecting the nervous system, while toluene and xylene can damage the nervous and reproductive systems. The main organs affected by these carcinogenic compounds are the lungs, liver, and kidneys. However, little attention has been paid to examining or summarizing the main published findings regarding biogas emissions. Furthermore, there is lack of knowledge about the environmental toxicological characteristics of most contaminants contained in landfill biogas. The main objectives of the present study are:

- Identification and quantification of BTEX emitted by landfill.
- Assessment and evaluation of the BTEX impact on human health.

Material and methods

Background and analytical method

The biogas was collected at the surface of final cover of a landfill in Saint Nicephore in Quebec, Canada (Fig. 1). This part where the measurement is done is inactive since 2002. A circular chamber (Fig. 2) was used to collect the biogas emitted at the surface. In this landfill, a passive methane oxidizing bio-cover (PMOB) was installed since 2006. The experimental PMOBs were constructed in 2006 on a parcel of the Saint Nicephore (Quebec, Canada) landfill that is inactive since 2002. This 60-ha site receives mainly municipal solid waste from Quebec cities. The experimental PMOBs measure 2.75 m (W), 9.75 m (L), with a depth of 1.20 m. PMOB-1B was constructed with the following materials: 0.8 m of substrate underlain by a 0.1-m-thick transitional layer (6.4-mm net gravel) and a 0.2-m-thick gas distribution layer (12.7-mm net gravel). More details about PMOB can be found in Lakhouit et al. (2014). BTEX maximum concentrations were then estimated. The collected emissions of biogas (~ 10 L) were sent for testing in an environmental engineering laboratory equipped with a gas chromatograph (GC; G1800A, Hewlett-Packard, Agilent Technologies, Mississauga, ON, Canada) that included an electron ionization detector (MS) and an HP-5 MS fused-silica column (30 m × 0.25 mm i.d., 0.25-mm film thickness, Hewlett-Packard, Agilent Technologies, Mississauga, ON, Canada). For extracting and concentrating the VOCs from the samples, a solid-phase micro-extraction (SPME) fibre (Carboxen/PDMS, 85 μm, SUPELCO, Bellefonte, PA, USA) was employed (Kleeberg et al. 2005). Prior to injecting it into each sample, the SPME fibre was thoroughly cleaned. Furthermore, prior to its first use, the fibre underwent a conditioning step in which it was placed inside a GC injector for 1 h at 300 °C. For each BTEX compound, calibration curves were made out of five concentrations in triplicate. The curves are known to be well suited to the SPME strategy (Ouyang and Pawliszyn 2006, 2008). Tedlar bags (10 and 44 L) were used as a foundation in constructing the samples for the calibration curves, with filtered air being added to fill the bag volume. The air was filtered using activated carbon, drierite, and silica gel. For BTEX's compounds (benzene, toluene, ethyl-benzene, and xylene), the detection limit of parts per billion (ppb) is estimated at 1.76, 38.13, 4.10, and 24.52, respectively, while the limit of quantification, also in ppb, is estimated at 5.85, 127.09, 13.66, and 81.75, respectively.

Estimation of rate inhalation (IR)

It is documented that biogas landfill contains BTEX compounds (Lakhouit et al. 2014; Ceron Breton et al. 2020; Herrero et al. 2020). In this section, the BTEX risk will be

Fig. 1 Study area in the landfill

assessed. The risk assessment of pollutants from landfills is becoming a major environmental issue (Herrero et al. 2020). The risk characterization is specifically determined by integrating information from the dose-response and exposure assessments. In the present study, the process is performed for the carcinogenic compounds BTEX. In Eq. 1, several parameters are estimated. The intake rate (IR) is calculated as follows:

$$IR = c\frac{CR.EF.ED}{BW.AT} \quad (1)$$

where

C average concentration of contaminant at exposure (mg/m3)
CR contact rate (in L/day, mg/day, or m^3/day)
EF exposure frequency (in days per year)
ED exposure duration (in years)
BW body weight (in kg)
AT period over which exposure is averaged (in days)

The EPA default values of these parameters are reported on Table 1.

Fig. 2 Circular chamber

Results and discussion

The IR was estimated and is reported in Table 2. The emitted biogas concentration ranges from below the limit of detection (BLD) to 3.76 mg m^{-3}. During the present study, the sampling period is taken place from June to September; the concentration of benzene was BLD. The difference in terms of BTEX concentration in this case is due to many factors, such as type of wastes, microbial activities, and layer of final cover (e.g. topsoil and vegetation), all of which can have an impact on biogas emissions (Lakhouit et al. 2014). Furthermore, physical, chemical, and physiological factors in the environment affect biodegradation of organic compounds, such as (i) availability of the compounds, (ii) availability of electron donors and acceptors, (iii) oxygen concentration, (iv) temperature, (v) pH, (vi) moisture, (vii) salinity, (viii) sorption of chemicals to particulate material, and (ix) concentration of the chemicals. For example, the amount of oxygen required for biodegradation of an organic fraction of MSW was estimated on the basis of stoichiometric equations and increased from 0.92 moles per 1 mole of waste at 20 °C to 1.6 moles at 42 °C within 96 h of the experiments (Liwarska-Bizukojc and Ledakowicz 2003). However, the emitted concentrations measured during the present study are in agreement with the literature and previous studies (Durmusoglu et al. 2010; Lakhouit et al. 2014, 2016).

Table 1 EPA default values for use in exposure assessment calculations for workers

Parameter	Case Worker
CR	30 m^3/day air inhalation
EF	250 days/year
ED	Actual event duration or 25 years, if chronic
BW	70 kg
AT (non-carcinogenic)	Same as ED
AT (Carcinogenic)	365 days/year × 70 years if

Table 2 Calculation of intake rate

BTEX	Concentration (mg m^{-3})	Intake rate (IR) (mg/kg day)
		Worker
Benzene	BLD*	N/A**
Toluene	3.76	0.39
Ethyl-benzene	2.60	0.27
Xylene	3.03	0.32

* = below the limit of quantification; ** = not applicable

In the scientific literature, VOC concentrations in landfill biogas range from 0.2 to 4500 mg m^{-3}. The concentration depends on the age, quantity, quality, and origin of the waste, which can vary from one landfill cell to another, as well as on the climatic conditions prevailing in the area where the landfill is installed (Durmusoglu et al. 2010).

One of the main objectives of the present study is to estimate the cancer risk of BTEX. A cancer risk assessment was performed using Eq. 2, as follows:

$$\text{Cancer risk} = \text{IR} * \text{SF} \quad (2)$$

where *I* is intake inhalation (mg/kg-day) and SF is slope factor $(\text{mg/kg-day})^{-1}$.

In the present study, SF = 0.0273 $(\text{mg/kg-day})^{-1}$. Table 3 shows the estimated cancer risk. Long-term exposure to BTEX affects the health of those who work in landfills. These risks are higher than the acceptable risk levels defined by the USEPA and the World Health Organization (WHO). According to the present study, we found an excess risk of cancer. The data from this study suggest that residents who lived near landfill sites may have been—and may continue to be—at excess risk of developing cancer as a direct result of biogas being emitted from the landfill site. Workers in landfill sites are also at a higher risk for developing cancer.

According to the literature (Ceron Breton et al. 2020), an acceptable environmental risk has been defined as the risk of an adverse effect of one chance in one million (10^{-6}) over a lifetime, whereas an unacceptable environmental risk has been defined as the risk of an adverse effect of one chance in one thousand (10^{-3}) over a lifetime. Additional investigations into VOCs in landfill sites should be conducted in order to refute or confirm these standard assumptions. For the BTEX compounds, benzene was the only BLD element, whereas the rest were detectable in a range where they could cause cancer to anyone working in or living near the landfill. Specifically, in this investigation, the estimated risk was clearly in the unacceptable range, although further investigation would be warranted to either refute or confirm these findings.

Landfills contain solid wastes and serve to prevent contamination between the waste and the surrounding environment (i.e. groundwater and atmosphere). In order to attenuate landfill emissions, a final cover is required. In fact, a final cover can be installed for a number of reasons, such as to prevent erosion and infiltration of precipitation, or for the control of odour and gas emissions. The efficiency of biogas collection ranges from 35 to 90%. Results of gas collection efficiency studies for various cover materials using flux box measurements are documented in Spokas et al. (2006). As the final cover plays an important role as a biogas emission reducer prior to venting to the atmosphere, final covers ultimately reduce human risk.

Table 3 Estimation of cancer risk

BTEX	Cancer risk Worker
Benzene	N/A*
Toluene	0.010
Ethyl-Benzene	0.007
Xylene	0.009

*= not applicable

Conclusions

The present study aims to study the impact of BTEX emitted by landfill. A landfill was chosen in Quebec. The landfill is situated near Drummondville City in Quebec. Landfills are a source of methane, carbon dioxide, unpleasant odours, and volatile organic compounds (VOCs). BTEX, a group of VOCs, presents an environmental and societal concern for landfill sites because they negatively affect the quality of life for workers at the landfill. During the present study, the emitted biogas at the surface of a final cover was collected and treated. A circular chamber was used for this purpose. An analytical method was developed to estimate the BTEX concentration. The limit of detection and quantification are estimated also. The biogas collected was treated by GC/MS and a fibre SMPE was used to absorb BTEX compounds. The concentrations of BTEX were in the BLD to 3.76 mg m^{-3}.

The intake rate was estimated, and its value was found to be in the range of 0.27 to 0.39 mg/kg-day. The present study constitutes a field documentation for the scientific communities. The main environmental and societal impacts of BTEX emissions are risks to workers' health associated with BTEX toxicity.

The cancer risk of BTEX estimated during this study was found to be in the range of 0.007 to 0.010. The estimated risk was clearly in the unacceptable range during our sampling period that is taken place from June to September. Considering these results, the major conclusion is that the evidence of adverse health outcomes for workers has thus far been insufficient and inconclusive. As this work showed, BTEX is released into the environment as a mixture, not a single compound. Hence, the toxicity and risk of BTEX should be investigated as a mixture and should also consider the two primary entrances of inhalation and ingestion. As a perspective of the present study, the authors suggest a sampling period over all the year if it is possible. In this landfill, there are no data concerning the biogas emissions from October to May. During this period, it is exceedingly difficult to have access to the inactive part of landfill where the PMOB is installed. The aerodynamic parameters, flora, and fauna existing in the landfill should be taken in account. The authors have no idea about the mass of solid waste because this part of landfill is inactive since 2002. In parallel with biogas emission and risk assessment of BTEX, leachate should be assessed and evaluated.

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Compliance with ethical standards

Conflict of interest The authors declare that there is no conflict of interest.

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