Environment International 97 (2016) 15–27

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Health risk impacts analysis of fugitive aromatic compounds emissions from the working face of a municipal solid waste landfill in China

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ARTICLE INFO

Article history:
Received 25 July 2016
Received in revised form 9 October 2016
Accepted 9 October 2016
Available online xxxx

Keywords:
Landfill
Aromatics
Emission factor
Uncertainty
Health risk assessment

ABSTRACT

Aromatic compounds (ACs) emitted from landfills have attracted a lot of attention of the public due to their adverse impacts on the environment and human health. This study assessed the health risk impacts of the fugitive ACs emitted from the working face of a municipal solid waste (MSW) landfill in China. The emission data was acquired by long-term in-situ samplings using a modified wind tunnel system. The uncertainty of aromatic emissions is determined by means of statistics and the emission factors were thus developed. Two scenarios, i.e. '*normal-case*' and '*worst-case*', were presented to evaluate the potential health risk in different weather conditions. For this typical large anaerobic landfill, toluene was the dominant species owing to its highest releasing rate ($3.40 \pm 3.79\ g \cdot m^{-2} \cdot d^{-1}$). Despite being of negligible non-carcinogenic risk, the ACs might bring carcinogenic risks to human in the nearby area. Ethylbenzene was the major health threat substance. The cumulative carcinogenic risk impact area is as far as ~1.5 km at downwind direction for the *normal-case* scenario, and even nearly 4 km for the *worst-case* scenario. Health risks of fugitive ACs emissions from active landfills should be concerned, especially for landfills which still receiving mixed MSW.

1. Introduction

Adverse health impact caused by landfills is an important and sensitive topic, which has received increasing social and environmental attention in terms of toxicity. Landfill workers and the nearby-living people are concerned about the potential health risk due to their direct exposure to the non-methane organic compounds (NMOCs) emitted from landfill through inhalation (Allen et al., 1997; Demir et al., 2012; Domingo and Nadal, 2009). However, in China and other Asian developing countries, the potential health effects of landfill gas (LFG) have not yet attracted sufficient attention due to lack of observation data.

Landfills are identified as a hazardous air pollutant source under the Urban Air Toxic Strategy by USEPA (1999). Although the amount of NMOCs is <1% (V/V) of the total landfill gas (LFG), their impacts on human health is not negligible. Aromatic compounds (ACs), major group of NMOCs in LFG, are considered to be one of the most hazardous types of air pollutants with neurotoxic, carcinogenic and teratogenic properties. Benzene, toluene, ethylbenzene, xylenes and styrene were dominant ACs in LFG, which have been documented under both aerobic and anaerobic conditions (Staley et al., 2006). Relative high ACs concentrations have also been reported from LFG in China, and among toluene was the most prevalent (Zou et al., 2003). Long-term exposure to benzene, toluene, xylenes and other ACs has been associated with a range of adverse health effects, including blood disorders and even cancer (World Health Organization, 2015).

Previous studies showed that the impact of LFG on health is within the acceptable levels (Davoli et al., 2010; Durmusoglu et al., 2010; Martí et al., 2014; Palmiotto et al., 2014). However, these studies were mainly conducted at closure area of landfills in developed countries. The situation of landfills in developing countries, such as China is quite different. On one hand, due to insufficient source separation, toxic wastes are simply mixed and landfilled with municipal solid waste (MSW), such as mothballs, pesticides, fragrance agents, toilet-deodorizers, solvents in paints and inks, foams, even herbicides and bactericides (Chen et al., 2010). These wastes can contain carcinogens and some of them are listed in the National Catalogue of Hazardous Wastes, which are significant sources of ACs. For example, benzene in leftover paints and pesticides are known to cause human cancer (IARC, 2016). On the other hand, MSW in China comprises high content of easily degradable organic wastes (>50%) and high moisture content (40–60%) leading to serious LFG fugitive emissions and air pollutions by NMOCs. Moreover, food waste is also an important source of ACs, as higher AC concentrations were found in higher fat content foods (Heikes et al., 1995). While cover layers in landfills could reduce large amount of NMOCs, working face is a significant source of fugitive NMOCs

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http://dx.doi.org/10.1016/j.envint.2016.10.010
0160-4120/

emissions (Scheutz et al., 2008). As the major component of NMOCs, ACs emitted from the working face through volatilization, biodegradation or chemical reaction processes. It should also be noted that the freshly mixed wastes would usually be exposed to the environment for days or even weeks prior to being well compacted and covered (Duan et al., 2014; Lu et al., 2015; Zhang et al., 2010). All these have made landfill sites be a significant source of carcinogenic substances in developing countries.

The reliability and accuracy of ACs emission data is crucial for risk assessment. Previously, a wind tunnel system was modified to measure the fugitive odor emission rates (Liu et al., 2015). Fieldwork has been done since 2012 for direct measurement of emission rates from the working face of a landfill in Beijing, China. According to their results, carcinogenic ACs, such as benzene, ethylbenzene and toluene, were found to be of relatively high concentrations. The potential health impacts therefore attracted more attention and the continuously in-situ sampling measurement was developed in 2014 for health risk assessment. In addition, ACs emission from landfills is a variable, complex process with rather uncertainty, so data analysis is necessitated to provide scientific and reliable results. Statistical analyses have been used to characterize the complexity of ACs emission (O'Connor, 2011).

The objective of the current study is to evaluate the potential exposure risk of ACs for residents living in the vicinity of an active MSW landfill. A few steps were taken: firstly, the probability distributions of ACs emission rates were determined and the emission factors were developed on the basis of in-situ sampling; then, the spatial distributions of ACs were calculated using a Gaussian dispersion model in a '*normal-case*' and '*worst-case*' scenario, respectively; finally, the potential health risk and the impact distance were assessed.

2. Materials and methods

The approaches adopted in this study involve site description, analytical measurements of emission rates, development of emission factors, characterization of the aromatic distribution using a Gaussian dispersion model, and chemical intake-based risk assessment.

2.1. Site description

The study was conducted on the working face of a landfill in Beijing, China (40.16 N, 116.35 E). Approximately 3000 $t \cdot d^{-1}$ of MSW is deposited in this flatland type landfill. The working face has an area of approximately 1000 m^2, where wastes are dumped and compacted daily (Fig. A1, Appendix A). A high percentage of organic matter are landfilled (~80%), such as kitchen waste, wood and paper.

2.2. Aromatic compounds emissions

A total of 14 ACs were detected from the samples (Table B1, Appendix B), the 6 of them, i.e. benzene, toluene, ethylbenzene, $p + m$-xylene, o-xylene (BTEX) and naphthalene were taken into account after comparisons with data provided by the International Agency for Research on Cancer (IRAC) and the integrated risk information system (IRIS). From a toxicological perspective, benzene has been classified as a known human carcinogen (Group 1) by the IRAC based on sufficient evidence, and chronic inhalation exposure would cause various blood disorders. Ethylbenzene and naphthalene are classified as possible human carcinogens (Group 2B). Although toluene, $p + m$-xylene and o-xylene are not classified as human carcinogens, they have significant non-carcinogenic toxic potentials. In addition, the presence of these compounds in landfill emissions has been well documented (Duan et al., 2014; Kim et al., 2008).

2.2.1. Gas sampling and analysis

A modified wind tunnel system reported in a previous study was adopted for the measurement of ACs emission rates from the working face (Liu et al., 2015). The system was firstly installed on the working face, where the fresh wastes were compacted and the compacted wastes were piled up. Nitrogen instead of the ambient air was used as carrier gas through the wind tunnel to prevent background contamination. According to the previous study, 19 $m^3 \cdot h^{-1}$ was an appropriate flow rate which can guarantee a steady and uniform flow performance inside the system. So, all the sampling campaigns were conducted at the fixed nitrogen flow of 19 $m^3 \cdot h^{-1}$, which equals a sweeping velocity inside the wind tunnel 0.26 $m \cdot s^{-1}$. After installation, the system was initially flushed with nitrogen for 60 s to 100 s until the steady flow rate state (19 $m^3 \cdot h^{-1}$) was achieved. Then air samples were withdrew from the outlet of the system to a 1 L multi-layer foil sampling bag (Dalian Delin Gas Packing Co., Ltd) with a SOC-01 sampler (National Key Laboratory of Odor Pollution Control of EPA-China, Tianjin), which was based on the lung principle.

Sampling campaigns were conducted on the working face of the landfill on selected days from May 2014 to January 2015. On each sampling day, air samples were collected every hour during the daytime (7 a.m. to 7 p.m.) and every 2 hours at night (7 p.m. to 7 a.m.) until the next morning. To minimize excessive heating and avoid direct exposure to sunlight, opaque sampling bags and a light-proof gas sampler (SOC-01, National Key Laboratory of Odor Pollution Control of EPA-China, Tianjin) were used during sampling. Gas bags were kept in an opaque box before measurement. A total of 124 valid samples were obtained, and meteorological parameters (i.e. temperature, humidity and air pressure) of the ambient environment at sampling spots were detected by thermometer, hygrometer and barometer, meteorological data on each sampling date were summarized in Table A1 (Appendix A).

All of the samples were analyzed within 24 h after collection. HAPSITE®ER (Inficon, East Syracuse, USA), a person-portable gas chromatography-mass spectrometer (GC–MS) equipped with a non-polar column (100% polydimethylsiloxane; 15 m × 0.25 mm ID × 1.0 μmdf), was used to quantitatively analyze ACs in the samples. Details of the quality control and quality assurance were provided in the Appendix B. ACs were quantified by an internal standard calibration procedure (internal standard contains 50 ppmv Bromopentafluorobenzene and 100 ppmv 1,3,5-trisbenzene balanced with high-pure nitrogen, Scott Specialty Gases, USA) that included the US NIST (National Institute of Standards and Technology) 98 Library. In addition, four levels (i.e., 0, 10, 20, 40 ppb) of TO-15 solution (Spectra Gases Inc. USA) were used as external calibration standards, and in all cases the linear fit with $R^2 > 0.99$ were achieved. The original data about the ACs concentrations in the samples were listed in Appendix C.

2.2.2. Emission rate computations

The emission rates (ER) of aromatics were calculated on the basis of their concentrations and the characteristics of the wind tunnel system, as the following:

$$ER = \frac{24 \times Q \times c}{B}, \quad (1)$$

where ER is the emission rate of the aromatics, $g \cdot m^{-2} \cdot d^{-1}$; Q is the nitrogen flushing flow rate, $m^3 \cdot h^{-1}$; c is the concentration of aromatics, $g \cdot m^{-3}$; and B is the base area of the wind tunnel, which was 0.1 m^2 in the current study.

2.2.3. Methodology development for ACs emission factors

Naturally, aromatic emissions are characteristically complex, random and uncertain. It is insufficient to describe highly variable aromatic emissions using simple averages ± standard deviations. So probability density functions (PDFs) were introduced to minimize the uncertainty problems. A PDF is a function that describes the relative likelihood of a random variable, the emission rate in this study, to take on a given value. In addition, considering the adverse impacts of ACs on human health, the 95% upper confidence limit (95th) emission rate was defined