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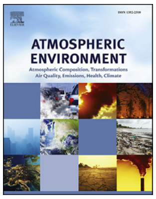

Aromatic compound emissions from municipal solid waste landfill: Emission factors and their impact on air pollution

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HIGHLIGHTS

- EFs of ACs from landfill in China were proposed based on statistical analyses.
- Use PDFs and CDFs to characterize the uncertainty of ACs emission rates.
- Toluene was the dominant AC with highest emission rate, 38.8 ± 43.0 $\mu g\ m^{-2}\ s^{-1}$.
- Air pollution caused by AC from landfill is less than other anthropogenic sources.

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ABSTRACT

Aromatic compounds (ACs) are major components of volatile organic compounds emitted from municipal solid waste (MSW) landfills. The ACs emissions from the working face of a landfill in Beijing were studied from 2014 to 2015 using a modified wind tunnel system. Emission factors (EFs) of fugitive ACs emissions from the working face of the landfill were proposed according to statistical analyses to cope with their uncertainty. And their impacts on air quality were assessed for the first time. Toluene was the dominant AC with an average emission rate of 38.8 ± 43.0 $\mu g\ m^{-2}\ s^{-1}$ (at a sweeping velocity of 0.26 $m\ s^{-1}$). An increasing trend in AC emission rates was observed from 12:00 to 18:00 and then peaked at 21:00 (314.3 $\mu g\ m^{-2}\ s^{-1}$). The probability density functions (PDFs) of AC emission rates could be classified into three distributions: Gaussian, log-normal, and logistic. EFs of ACs from the working face of the landfill were proposed according to the 95th percentile cumulative emission rates and the wind effects on ACs emissions. The annual ozone formation and secondary organic aerosol formation potential caused by AC emissions from landfills in Beijing were estimated to be 8.86×10^5 kg $year^{-1}$ and 3.46×10^4 kg $year^{-1}$, respectively. Toluene, $m + p$-xylene, and 1,3,5-trimethylbenzene were the most significant contributors to air pollution. Although ACs pollutions from landfills accounts for less percentage (~0.1%) compared with other anthropogenic sources, their fugitive emissions which cannot be controlled efficiently deserve more attention and further investigation.

1. Introduction

Numerous volatile organic compounds (VOCs) arise from volatile compounds in waste and those formed during waste decomposition (Tian et al., 2013). Aromatic compound (AC) emissions from landfills are among the major VOC sources aside from industries and vehicles, which significantly contribute to the current atmospheric VOC concentration (Zou et al., 2003). Aside from having neurotoxic, carcinogenic, and teratogenic properties, ACs have been traditionally considered to be the most important secondary organic aerosol (SOA) precursors in the troposphere (Durmusoglu et al., 2010; Kumar et al., 2011; Yang et al., 2014). Reportedly, the increasing PM 2.5 pollution events are largely

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driven by SOA formation (Huang et al., 2014). AC emissions from the working face in landfills, that receive fresh waste daily, are more difficult to estimate and control than industrial or mobile emissions (Carlos et al., 2012; Duan et al., 2014; Gallego et al., 2014). Although AC identification and quantification in urban air have been extensively studied (Ding et al., 2012; Fang et al., 2012; Kim and Kim, 2002), AC emission characteristics from the working face of municipal solid waste (MSW) landfills and their adverse impacts on air quality are seldom reported (Kumar et al., 2011; Song et al., 2007; Yang et al., 2014).

An emission factor (EF) is a representative value that relates the quantity of a pollutant released to the atmosphere with an activity associated with the release of that pollutant. USEPA (2012) provides non-methane organic compound (NMOC) EFs from MSW landfills in The AP-42 Compilation of Air Pollutant Emission Factors (AP-42), where the total NMOC in landfill gas (LFG) ranges from 31 ppmv to over 5387 ppmv, with an average of 838 ppmv. European Environment Agency (UK, 2013) gives a default EF of NMOC for the source of solid waste disposal on land in Europe, with a value of 5.7 g m^{-3} landfill gas. However, no EFs of VOCs are available for landfill fugitive emission source in China. Moreover, it is inappropriate to evaluate Chinese landfill VOCs emissions using the database from USA or EU, because wastes in China are characterized by high content of food waste (60%) (Yang et al., 2014), while in the USA and Europe food waste usually constitutes between 30 and 40% of the total MSW (Tugov, 2015; USEPA, 2015).

Emission rate is fundamental information to evaluate EFs. To measure fugitive gas area emissions, the wind tunnel system, originally developed by the University of New South Wales (Jiang et al., 1995), has been adopted. The system is recommended as it can capture fugitive gas emissions and simulate the process of wind blowing through the surface at the same time. In addition, the measured emission rates are particularly appropriate to be used for dispersion model (Capelli et al., 2009; Hudson and Ayoko, 2009). The system has been modified by Liu et al. (2015) for assessing VOCs emission rates from the working face of a landfill.

Previously, EFs are usually calculated based on the simply average value of observation data with acceptable quality (Sironi et al., 2005). However, AC emission from the working face of a landfill is a high variable and uncertain process, which might be caused by the variances in waste compositions, meteorological conditions, and operation modes. To evaluate EFs more specifically and reasonable, the statistical analysis of probability density function (PDF) might be advisable for the estimation of EFs. PDF is a function that describes the relative likelihood for a random variable to adopt a specific value.

This study attempts to determine emission factors of aromatic compounds from MSW landfill and afterword assess the impacts on air pollution. A modified wind tunnel system is introduced to measure AC emission rates from the working face of a landfill, and further analysis is performed using a gas chromatography–mass spectrometry (GC–MS) system. EFs are evaluated based on the PDFs of AC emission rates. Subsequently, the potential of ozone and SOA formations from AC emissions are calculated. The current study comes up with a novel approach to help determine the complex, uncertain and random AC emissions from the landfill.

2. Materials and methods

2.1. Site description

The study was conducted on the working face of a MSW landfill in Beijing, China (40.16 N, 116.35 E). It was a typical landfill in flat lands, where approximately 3000 t d^{-1} of MSW was deposited. The working face comprised an area of approximately 1000 m^2, where wastes were dumped and compacted daily (Fig. A1, Supporting information). The seasonal variation of waste composition was discussed by Duan et al. (2014). The landfill was highly composed of organic matter, and food waste was the main component accounting for 40%–60% (by weight). The comprising of paper and plastics was higher in winter than summer.

2.2. Gas sampling

A modified wind tunnel system was adopted to measure the emission rates of ACs from the working face of the landfill. The system was developed and tested by Liu et al. (2015) previously. The wind tunnel system was firstly installed on the working face, where compacted fresh wastes were exposed to the environment. The main chamber of the system was partly inserted into the garbage and leveled with the working face. Ambient air was usually introduced as carrier gas through the wind tunnel, but this is inappropriate in the current study, as the air in landfill is severe contaminated, which would cause great background noise. Hence, nitrogen was used as carrier gas through the wind tunnel, supplied by nitrogen cylinders. After installation, the system was initially flushed with nitrogen for 60 s–100 s (with a volume of 300 L–500 L) until a steady flow rate (19 m^3 h^{-1}) state was achieved. Then air samples were collected from the outlet with a SOC-01 sampler, which was based on the lung principle. Gas samples were withdrew to a 1 L multi-layer foil sampling bag (Dalian Delin Gas Packing Co., Ltd) through a polytetrafluoroethylene sampling tube. All sampling campaigns were conducted at a fixed nitrogen flushing rate of 19 m^3 h^{-1}, where the flow performance inside the wind tunnel system has been proved to be steady and uniform. So the sweeping velocity inside the wind tunnel equals 0.26 $m \cdot s^{-1}$. The details of the system were described by Liu et al. (2015) and sampling procedure by SOC-01 sampler was discussed by Duan et al. (2014).

Sampling campaigns were conducted at the aforementioned landfill on 7 days from May 2014 to January 2015. On each sampling day, air samples were collected every hour during the day, like 8 a.m. to 6 p.m., and every 2 h at night, like 7 p.m. to 7 a.m. next morning. Direct exposures to sunlight before, during, and after sampling were carefully avoided to minimize excessive heating of the samples. A total of 124 valid samples were obtained, and meteorological information (i.e., temperature, humidity, and air pressure) of the ambient environment at sampling spots was recorded while sampling (see Table A1, Supporting information).

2.3. Gas chromatography–mass spectrometry (GC-MS) analysis

The samples were analyzed within 24 h after collection. HAPSITE®ER (Inficon, East Syracuse, USA), a person-portable 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 gas samples. The heating program was described in a previous study (Liu et al., 2015). ACs were identified on the basis of their retention time, target, and qualifier ions. Identified compounds were quantified using the internal standard calibration procedure, including the US NIST (National Institute of Standards and Technology) 98 Library and the US Environmental Protection Agency standard solution (EPA TO-15) (US, 1999). Three levels (i.e. 10, 20, 40 ppb) of TO-15 solution were used as calibration standards, and in all cases linear fit was good with $r^2 > 0.99$.

2.4. Data analysis

2.4.1. Emission rate computations

Emission rate was defined as the amount of AC emission per unit