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Original scientific paper

Health Risk Assesssment of Benzen, Toluene and Xylene at a Landfill

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ABSTRACT: The municipal solid waste (MSW) landfill is recognized as an anthropogenic source of air pollutants that can have a negative impact on human health and the environment. Workers who work at the MSW landfill may be exposed to risk due to the inhalation of substances such as volatile organic compounds (VOCs). Although VOCs account for <1% in landfill gas, they are important because of the high level of toxicity associated with them. Regular monitoring of air quality and risk assessment provides important information in protecting the health of workers at the landfill. This study focuses on a health risk assessment related to VOCs (benzene, toluene and xylene) exposure via inhalation for workers at a landfill Banja Luka, Republic of Srpska, Bosnia and Herzegovina. Additionally, cancer risk and non-cancer risk of benzene, toluene and xylene of workers indicated that occupational exposures were above recommended standard. This implies that landfill workers are exposed to a significant health risk associated with inhalation exposure to VOCs

Keywords: municipal solid waste, landfill, risk assessment, VOCs, human health.

INTRODUCTION

Landfilling is still one of the most widespread ways of municipal solid waste (MSW) management in the world (Luo et al., 2020). Landfill products such as landfill gas, leachate and odors can pose a risk to human health and the environment. (Przydatek & Kanownik, 2019). Landfill gas is generated by numerous processes in the body of the landfill, mainly organic waste decomposition processes (Pan et al., 2018). Landfill gas consists primarily of carbon dioxide (CO_2) and methane (CH_4), including some trace components such as hydrogen sulfide, hydrogen, carbon monoxide and nonmethane organic compounds (NMOCs) (Purmessur & Surroop, 2019). Landfill gas is a biogas consisting of approximately 45–60% (v/v) methane (CH_4) and 40–60% (v/v) carbon dioxide (CO_2), minor components consisting of oxygen (0.1–1%), hydrogen (0–0.2%), nitrogen (2–5%), carbon monoxide (0–0.2%), sulfides (0–1%), ammonia (0.1–1%) and a large number of trace components (0.01–0.6%). In landfill gas, the presence of 63 types of NMVOCs was detected in trace amounts in the range of 1–10,000 μg/L (Manheim et al., 2021). The NMVOCs include aliphatic compounds (alkanes and alkenes), aromatic hydrocarbons (benzene, toluene, ethylbenzene, xylene, etc.), halogenates (dichlorofluoromethane, vinyl chloride, etc.), hydrocarbons and alcohols (ethanol, methanol, etc.) (Pecorini et al., 2020). Volatile organic compounds (VOCs) comprise 39% of the NMVOCs at MSW landfill. VOCs are one of the major air pollutants (benzene, toluene, xylene) due to their malodorous and hazardous properties and can contribute to global warming, stratospheric ozone depletion and tropospheric ozone formation (Durmusoglu et al., 2010). The environmental significance of VOCs is well known due to their potential role as carcinogenic and mutagenic compounds and in the formation of photooxidants (Jayawardhana et al., 2019).

Benzene is a proven carcinogen, classified by International Agency for Research on Cancer (IARC) as an agent carcinogenic to humans (group 1). Toluene and xylene are not classifiable in regard to their carcinogenicity to humans (group 3) (Zdeb & Lebiocka, 2016). Benzene is a well-known carcinogenic component of landfill gas. Exposure to high concentrations of benzene can cause leukemia, neurological effects, skin problems, respiratory problems and eye problems. A high risk of leukemia can be expected when exposed to concentrations of benzene equal to or above 160 mg/m^3/year. Potential health effects depend on both concentration and period of exposure (Moolla et al., 2013). Toluene is applicable in the industry as a solvent. It can cause fatigue, dizziness, nausea, hearing loss, reduced color clarity of vision, and even blindness in some cases at low to mid-level. Breathing high levels of toluene can cause unconsciousness and even death. Long-term exposure to toluene is usually caused by neurological diseases and visual disturbances. Xylene affects the nervous system, and the symptoms are headache, dizziness, vomiting, and nausea (Kaydi et al., 2022). Inhalation of toluene and xylene causes headache, vertigo and inflammation of the mucous membrane (Peng et al., 2015).

The subject of determining the probability of an undesirable health effect from exposure to hazards is called "risk assessment". Risk assessment is one of the fastest evolving tools to evaluate the impact of the hazards on human health and to determine the level of treatment required to solve a specific environmental problem (Durmusoglu et al., 2010).

Many studies have been conducted in the world that investigated the content of benzene, toluene and xylene in landfill gas, as well as the assessment of the risk of these pollutants to human health. The studies that have been arranged so far included health risk assessment of benzene, toluene, ethylbenzene and xylene (BTEX) in ambient air on landfill and occupational health risk assessment BTEX on landfill or landfill gas (de Sá Borba et al., 2017; Durmusoglu et al., 2010; Khademi et al., 2022; Liu et al., 2022; Moola et al., 2013; Yaghmaien et al., 2019). In the Republic of Srpska (RS), Bosnia and Herzegovina (B&H), there have been no studies that dealt with the concentrations of benzene, toluene and xylene in the landfill gas, i.e. the ambient air of the landfill, as well as the impact of these pollutants on the health of workers and the population around the landfill.

Two studies in the area of B&H dealt with the presence of BTEX in the sediments of Sava and Bosna rivers (Medunić, & Šmit, 2016) and in the ambient air in school classrooms. (Beregszaszi et al., 2013). Benzene concentrations were determined near the coke plants in the industrial areas of Zenica and Lukavac (Đozić et al., 2016) and benzene and products of photochemical degradation of benzene in presence of other air pollutants were identified in Tuzla area (Marić et al., 2008). Ilić et al., (2020) measured the concentration of chemical hazards in the clinical hospital "St. Luke the Apostle" in Doboj (RS), among other benzene, toluene and xylene. The results of the measuring concentration of chemical hazards in the hospital showed that contamination with different harmful chemicals.

In this study the focus is on the landfill in Banja Luka, RS, B&H, and on determining the concentration of benzene, toluene and xylene. The aim of this work is (1) identification and quantification of benzene, toluene and xylene emitted from landfill gas, and (2) assessment and evaluation of the impact of these compounds on the health of landfill workers.

MATERIALS AND METHODS

Study area

Landfilling of waste in B&H is still the most important segment in waste management. In addition to existing regional and sanitary landfills, waste is still disposed of in illegal and unregulated landfills. Waste management in the Banja Luka region is based on waste disposal at the sanitary regional landfill located in the City of Banja Luka (Bjelić et al., 2022). The waste that is deposited at this landfill is brought from three cities (Banja Luka, Gradiška and Laktaši) and five municipalities (Srbac, Čelinac, Kotor Varoš, Kneževo, Prnjavor) (Figure 1). The total number of inhabitants in the region is 370,329.

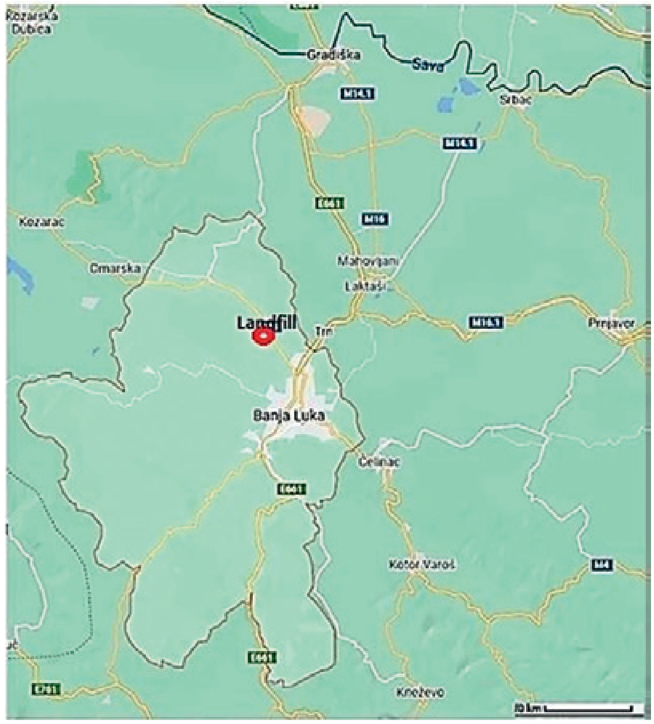


Figure 1. The position of cities and municipalities in relation to the Banja Luka landfill

MSW has been deposited at this landfill since 1976. Since the waste separation system has not yet been established, mixed MSW and non-hazardous industrial waste are disposed of at this landfill. The total amount of disposed waste in 2020, 2021 and 2022 was 121,086.71 tons, 125,487.94 tons, and 128,517.07 tons, respectively. The amount of disposed waste per inhabitant is 327 kg/year (0.90 kg/day) in 2020, or 339 kg/year (0.93 kg/day) in 2021 and 347
kg/year (0.95 kg/day) in 2022.

In the MSW at the Banja Luka landfill, the most dominant organic fraction is waste from the kitchen and garden waste such as food, fruit and vegetable remains and green waste from the garden (about 31%). Paper and cardboard with a share of 26% are in the second place by mass share in MSW. Other fractions of waste have shares in MSW below 10%. Fractions of waste classified as packaging waste and waste that could be separated and recycled such as paper, cardboard, metal, cans, glass, plastic, and PET make up about 45% of municipal waste (Euro- inspekt, 2016).

Sampling and analysis procedure

Air samples at the Banja Luka landfill were taken from two measurement locations: (1) on the surface of the landfill, i.e. on the current surface where the MSW is deposited, and (2) at the entrance to the landfill, which is about 200 m as the crow flies from the disposal zone. This sampling and measurement was carried out in the summer and winter periods of 2018 and 2021.

The concentrations of benzene, toluene and xylene were analyzed using gas chromatography according to BAS EN ISO/IEC 17025:2018 (General requirements for the competence of testing and calibration laboratories). The Clarus 680 Perkin Elmer gas chromatograph has the following characteristics: temperature range 20ºC-250ºC, maximum heating rate 140ºC/min, Elite-Wax column, length 60 m, ID 0.32, DF

0.5. For all compounds the detection limit for the concentration was <0.4 $\mu g \cdot m^{-3}$. Air sampling and analysis were performed by an accredited laboratory.

Risk calculation

Determining the extent of the negative effects of VOCs (benzene, toluene and xylene) on workers, who are exposed to higher concentration of these compounds for a longer period, is essential given the adverse health effects of these compounds. Risk assessment associated with chronic exposure to toxic compounds is an accepted method to assess the cancerous effects and non-cancerous risk of hazardous substances (Mo et al., 2021).

Life cancer risk (LCR) for benzene is calculated based on the following:

$$LCR = I \text{ x } CPF \qquad (1)$$

where:

I - daily intake (mg kg^{-1} day^{-1}),

CPF – carcinogen potency factor. CPF for benzene is 0.029 mg kg^{-1} day^{-1}.

Non-carcinogenic risk is determined in terms of hazard ratio (HR). The HRs for toluene and xylene are calculated based on the following:

$$HR = \frac{I}{RfD} \qquad (2)$$

RfD – reference dose. RfDs for toluene and xylene are 1.43 and 0.029 mg kg^{-1} day^{-1}, respectively.

Various factors that should be considered in determining the intake of contaminants include considerations of frequency, duration of exposure, and the body weight of the receptor. The intake rate is calculated as follows:

$$I = \frac{C \text{ x } CF \text{ x } IR \text{ x } EF \text{ x } ED}{BW \text{ x } AT} \qquad (3)$$

where:

C – contaminant concentration (μg m^{-3}); CF – conversion factor (mg μg^{-1});

IR – inhalation rate (m^3 day^{-1});

EF – exposure frequency (day $year^{-1}$); ED – exposure duration (year);

BW – body weight (kg), AT – averaging time (day).

CF is the concentration conversion (mg/µg = 0.001 or 1 µg) factor. IR for adults is 20 $m^3 \cdot day^{-1}$. The EF and ED are other important parameters which must be defined for calculating daily intake. In this study the risk was calculated for workers in landfill site with the daily mean laboring duration of 8 h (except Sundays). EF is 74 days $year^{-1}$ and work lifetime (ED) is assumed to be 20 years. BW is the average body weight (70 kg for male) and AT value of 70 years (or 25,500 days) can be used.

An LCR greater than 1×10^{-4} indicates a maximum individual risk for cancer. Normally the LCR should be below 1×10^{-6} (Cerón Bretón et al., 2020). An HR below 1 is considered to be a negligible risk (Hazrati et al., 2016).

RESULTS AND DISCUSSIONS

Benzene, toluene and xylene concentration

Table 1 presents the concentrations of benzene, toluene and xylene measured at the Banja Luka landfill in July and December 2018 and 2021 on the surface of the landfill and at the entrance of the landfill (ambient air), which is about 200 m away from the waste disposal zone. The active landfill is still receiving mixed MSW with no temporary soil cover for days.

Table 1. Concentration ranges of benzene, toluene and xylene on Banja Luka landfill (in mg/m^3)

	Location							
Parameter	**In landfill surface**				**In the air around the landfill**			
Benzene (CAS number 71-43-2)								
	Max	Min	Mean	Std.	Max	Min	Mean	Std.
2018 (July)	3.36	0.96	3.27	2.78	0.89	0.03	0.39	0.45
2018 (December)	4.03	1.47	2.34	1.46	0.78	0.10	0.48	0.34
2021 (July)	3.84	0.84	1.87	1.91	0.65	0.09	0.33	0.29
2021 (December)	3.39	0.89	2.07	1.26	0.39	0.01	0.20	0.19
Sum	4.03	0.84	2.89	1.62	0.89	0.01	0.38	0.33
Toluene (CAS number 108-88-3)								
2018 (July)	30.58	11.68	21.40	5.52	1.35	0.54	0.96	0.41
2018 (December)	52.43	27.41	31.43	4.73	0.98	0.36	0.61	0.33
2021 (July)	38.74	21.84	26.53	6.03	0.89	0.05	0.57	0.46
2021 (December)	36.92	19.39	20.40	7.34	1.15	0.50	0.85	0.33
Sum	30.58	11.68	25.08	5.69.	1.35	0.05	0.73	0.35
Xylene (CAS number 106-42-3)								
2018 (July)	4.04	1.29	2.18	1.82	0.69	0.41	0.59	0.16
2018 (December)	2.21	0.94	1.84	0.87	0.71	0.30	0.55	0.22
2021 (July)	2.58	1.03	1.41	0.61	0.74	0.38	0.55	0.18
2021 (December)	3.04	1.35	1.95	0.97	0.66	0.39	0.55	0.14
Sum	4.04	0.94	1.78	0.90	0.74	0.30	0.56	0.18

The minimum concentration of benzene on the surface of the deposited waste was 0.84 mg·m^{-3} and in the ambient air 0.01 mg·m^{-3}. The highest concentration of benzene during the measurement on the surface of the landfill was 4.03 mg·m^{-3} while in the ambient air at the entrance of the landfill it was 0.89 mg·m^{-3}. Apart from that, several countries Turkey, Mexico, and Malaysia have reported benzene concentrations in landfill gas, respectively, as follows 4.0 –6.7 mg·m^{-3}, 38 µg·m^{-3}, 1,191.5 mg·m^{-3} (Sevimoğlu & Tansel, 2013; Araiza-Aguilar et al., 2019; Kamarrudin et al., 2013). According to the Rulebook on preventive measures for safe and healthy work when exposed to chemical substances (Official Gazette of the Republic of Srpska number: 4/20), the maximum allowed concentration of benzene is 3.25 mg·m^{-3}. When measuring the air quality on the surface of the landfill, the maximum measured concentrations of benzene were above the permitted values.

The maximum concentration of toluene on the surface of the deposited waste was 30.58 mg·m^{-3}, and in the ambient air at the entrance to the landfill, the maximum concentration of toluene was 1.35 mg·m^{-3}. The minimum concentration of toluene on the surface of the deposited waste was 11.68 mg·m^{-3} and in the ambient air 0.05 mg·m^{-3}. In study, Turkey, Canada, and Iran have reported toluene concentrations in landfill gas, respectively, as follows 1.51-47.42 µg·m^{-3}, 3.76 mg·m^{-3}, 3.8-16.1 µg·m^{-3} (Dincer et al., 2006; Lakhouit & Alsulami, 2020; Yousefian et al., 2020). According to the Rulebook on preventive measures for safe and healthy work when exposed to chemical substances (Official Gazette of the Republic of Srpska number: 4/20), the maximum allowed concentration of toluene is 192 mg·m^{-3}. When measuring the air quality on the surface of the landfill and the ambient air, the concentration of toluene was below the permitted values.

The minimum concentration of xylene was 0.30 mg·m^{-3} in ambient air, while the maximum concentration of xylene was4.04 mg·m^{-3}. At the Iranian landfill, the concentration of xylene in the air around the landfill was in the range of 0.09-27.17 mg·m^{-3} and in the biogas pipe 0.03-23.44 mg·m^{-3} (Khademi et al., 2022). According to the Rulebook on preventive measures for safe and healthy work when exposed to chemical substances (Official Gazette of the Republic of Srpska number: 4/20), the maximum allowed concentration of xylene is 221 mg·m^{-3}. When measuring the air quality on the surface of the landfill and the ambient air, the concentration of xylene was below the permitted values.

In previous research at similar landfills, aromatic compounds (used as solvents in many products) are found in mixed municipal waste, and can be produced and released during aerobic or anaerobic decomposition, where benzene, toluene and xylene were detected on compacted and covered open cells landfill (Cheng et al., 2019; Yousefian et al., 2020). Each landfill may have a unique emission pattern due to differences in climate, topography, landfill characteristics, landfill operating conditions, etc. These may affect the benzene, toluene, and xylene profiles in each landfill. Ratios toluene:benzene (T:B) are most often used to estimate emissions from vehicles and industrial sources (Dave et al., 2020). The mean T:B ratios measured in this study in surface landfill are calculated as 9. High T:B ratios (T:B>10) have been reported commonly in many industrialized regions of the cities (Durmusoglu et al., 2010).

Cancer risk and hazard risk calculations

Cancer risk less than or equal to 1 to 1,000,000 (=1.0 x 10^{-6}) is considered insignificant or ineffective. On the other hand, projected risk greater than or equal to 1 to 1,000 (=1.0 x 10^{-3}) was defined as a warning for significant risk. The risk level of 1.0 x 10^{-4} was recently considered acceptable.

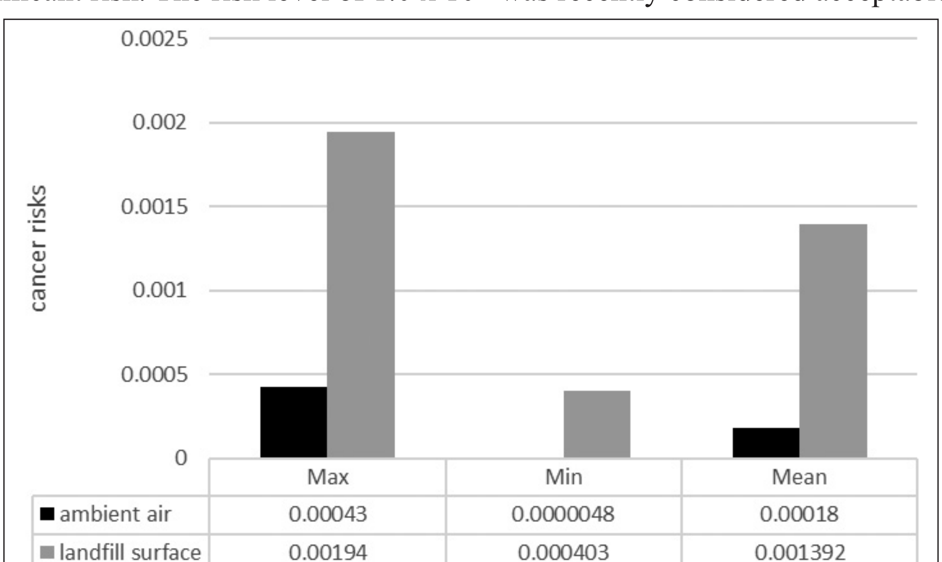
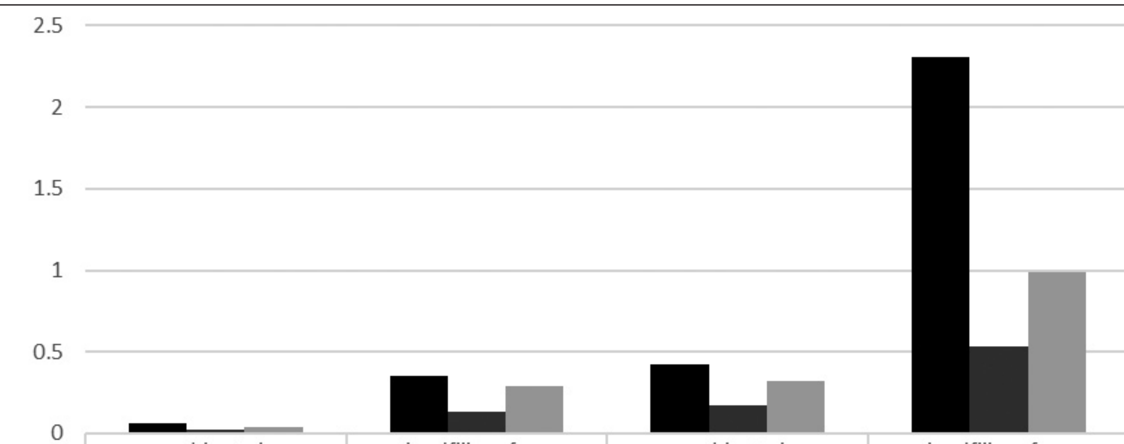


Figure 2. Cancer risks benzene in ambient air and landfill surface

The minimum, average and maximum benzene related cancer risks were predicted to be 0.4 x 10^{-3}, 1.39 x 10^{-3} and 1.9 x 10^{-3} in landfill surface, respectively. Here, both average and maximum carcinogen risk for benzene are exceeded 1.0 x 10^{-3}. Hence, the benzene cancer risk poses a significant risk at surface landfill site. The average benzene cancer risk suggests that 1,390 people will be at risk of cancer per million people in exposed population (Figure 2).

The minimum, average and maximum benzene related cancer risks were predicted to be 0.005 x 10^{-3}, 0.18 x 10^{-3} and 0.4 x 10^{-3} in ambient air, respectively. Here, minimum, average and maximum carcinogen risk for benzene are less than 1.0 x 10^{-3}. The average benzene cancer risk suggests that 180 people will be at risk of cancer per million people in exposed population. In the monograph of IARC, it is stated that there is no safe level of benzene, i.e. the carcinogenic component of benzene and its respective value should be zero (IARC, 2016).

For non-carcinogenic health effects, a hazard ratio was used to determine the risk. In order to evaluate the non-carcinogenic effects of the chemicals, it is common to consider the risk to be negligible if the HR is less than or equal to 1 (Hazrati et al., 2016). HR is determined and calculated for toluene and xylene.



	toluene		xylene	
	ambient air	landfill surface	ambient air	landfill surface
Max	0.06	0.35	0.42	2.31
Min	0.02	0.13	0.17	0.53
Mean	0.04	0.29	0.32	0.99

Figure 3. Hazard ratio for toluene and xylene in ambient air and landfill surface

The average HRs for toluene and xylene in landfill surface were predicted to be 0.29 and 0.99, respectively (Figure 3). The toluene and xylene average non-carcinogenic risks predicted at the landfill surface and ambient air are negligible because all of the HRs are less than one. On the other hand, the maximum HRs for toluene and xylene in landfill surface were 0.35 and 2.31, respectively. While the maximum non-carcinogenic risk for toluene is still much lower than 1.0, the risk for xylene is more than 1.0, thereby indicating an adverse health effect. However, this maximum value for xylene represents significant overestimate of potential non-carcinogenic risks. In the case of exposure to two or more substances that may be potentially hazardous to health, the combined effects of the independent effects of each of them should be taken into account.

To reduce the exposure of workers on landfill surface, the following policy is recommended (Yousefian et al., 2020): (1) expanding and improving the ventilation system, (2) adopting engineering control systems for the collection of gas, (3) implementing suitable working schedules based on exposure rates considering employee turnover and reducing the working time, and (4) minimizing health risks by using appropriate personal protective equipment for on-site workers such as face masks.

CONCLUSONS

Landfills represent one of the most important industrial sources of air pollution. These emissions may contain pollutants potentially dangerous for human health, and for this reason it is important to investigate the exposure risk for workers on landfill. Higher concentrations of benzene, toluene and xylene were found in landfill surface. The carcinogenic risk for occupational exposure to benzene was between 0.4 x 10^{-3} and 1.9 x 10^{-3}. The estimated occupational cancer risks demonstrated probable to significant risks at surface landfill site. The average HRs for toluene and xylene in landfill surface were predicted to be 0.29 and 0.99, respectively. The maximum values for xylene represent significant overestimates of potential non-carcinogenic risks.

With regard to cancer and non-cancer risks of benzene, toluene and xylene for workers on MSW landfill, the focus in prevention and protection of the health of workers should be on:

(1) minimizing exposure by reducing the exposure time of workers, (2) using appropriate personal protective equipment, (3) improvement of waste management, starting with methods of collection, recycling, treatment and final disposal at the landfill.

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