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Research paper

Characterizing volatile organic compounds in leachate from Gohagoda municipal solid waste dumpsite, Sri Lanka

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ABSTRACT

Fate and transport of volatile organic compounds (VOCs) in landfill leachate causes environmental pollution and human health concern. These VOCs can directly be emitted to the atmosphere and also can end up in leachate plume from open solid waste dumpsites. However, no data exists on the VOCs in landfill leachate in Sri Lanka. Hence, the purpose of this study was to assess the variation of VOCs in landfill leachate at the Gohagoda Municipal Solid Waste (MSW) dumpsite, Kandy, Sri Lanka in a dry (Spring) and a wet (Autumn) month. A list of the key offensive VOCs covering 13 chemicals in leachate was quantified by using a head space–gas-chromatography–mass-spectrometry (HS-GC–MS). Results indicated that benzene and toluene were most commonly observed in both sampling periods, ranging from 1.78–21.7 and 1.73–20.2 $\mu g\,L^{-1}$, respectively. 4-Isopropyltoluene had the highest concentrations of the VOCs investigated in both sampling periods (38.0 and 129 $\mu g\,L^{-1}$ in spring and autumn, respectively). Totally, 9 different VOCs out of 13 listed were detected in the landfill leachate. In comparison, the number of VOCs and total VOC concentrations were higher at all sampling locations in spring except GSV1 sampling point. Thereby, it appears that a time series analysis of VOCs during the dry and wet months may provide a better understanding of the degree and risk of pollution of the Mahaweli River water.

1. Introduction

Open dumping of Municipal Solid Waste (MSW) is the most common method of solid waste disposal in many developing countries due to the low budget for waste disposal and non-availability of skilled labor (Ali et al., 2014). An open dumpsite is a disposal site of unplanned heaps of uncovered wastes disposed of with no protection from environmental contamination (Joseph et al., 2003). Those are often burnt and surrounded by pools of stagnated polluted water (Ali et al., 2014). MSW dumpsites contain a variety of pollutants such as heavy metals, nutrients, dissolved solids and organic carbon, various organic pollutants including xenobiotic compounds, pesticides, hydrocarbons, etc. (Kjeldsen et al., 2002; Christensen et al., 2001). VOCs, particularly, benzene, toluene, ethyl-benzene and xylene isomers (collectively called BTEX), are found in many items in the household and industrial wastes (i.e., paints, cleaners, paint thinners, finger nail polish remover, etc.) that are directly disposed of in MSW dumpsites (Kjeldsen et al., 2002). These pollutants either end up in the atmosphere or in the leachate, the fluid percolating from landfills that are generated from liquids present in the waste and water from outside percolating though the waste. In most developing countries, the leachate from the MSW dumpsites directly flows to the surface water bodies with zero treatment (Wijesekara et al., 2014). Therefore, landfill leachate characterization is exceedingly important for the development of compatible strategy and remediation technique (El-Fadel et al., 2002).

Heavy metals and organic pollutants in landfill leachates have been studied thoroughly and presented in detail (Renou et al., 2008). However, the VOCs, which is one of the highly toxic carcinogenic compounds, where the toxicity have not been fully investigated. US Environmental Protection Agency (USEPA) defined the VOCs as stable products exhibiting the vapor pressure above 0.1 mmHg (13.332 Pa) at ambient conditions (Giaya et al., 2000). The different composition of VOCs rather than the single

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Table 1
Benzene and few other VOC concentrations recorded in landfill sites from different countries.

Country	VOCs (μg/L)				Reference
	Benzene	Toluene	Ethyl benzene	Xylene	
Korea	32 ± 61.8	259 ± 287	–	–	Kim et al. (2006)
Italy	–	1.62	–	–	Davoli et al. (2003)
China	0.73	0.12	–	–	Zou et al. (2003)
Denmark	1.5	615	383	236	Scheutz et al. (2004)
USA	12	–	–	–	Wood and Porter (1987)
USA	17–540	7.5–600	12–820	12–170	Sabel and Clark (1984)
Hawai	0.47	–	–	–	Mournighan et al. (2007)
Switzerland	8.8–128	–	–	–	Riediker et al. (2000)
USA	0–10	0–3.5	0–0.1	0–1	Eganhouse et al. (2001)
USA	19	280–1600	100–250	–	Sabel and Clark (1984)
Spain	1.1–572	3.8–48	1.7–20	3.7–38	Först et al. (1989)
USA Wisconsin	1.4–220	1.2–610	1.4–180	2.5–240	Friedman (1988)
Sri Lanka	1.78–21.7	1.73–20.2	1.30–6.36	1.45–6.76	This study

compound, the dose and the time can lead to a diverse impact on human health. Health effects from VOCs can be distinguished to acute, chronic not including cancer and cancerous (Cohen et al., 2005; Künzlia and Tagerb, 2005). Some VOCs are present in trace concentrations but the potential risks of these contaminants cannot be ignored due to their adverse effects on human health (Kreith and Tchobanoglous, 1994). The VOCs are found as components of landfill gas and leachates formed by degradation and volatilization of organic materials deposited in the landfill (Table 1). A part of these VOCs ends up in leachate that forms as water percolates through the waste mass (Chiriac et al., 2007). The VOCs such as benzene, toluene, ethylbenzene, and xylenes are the major pollutants found in groundwater and are common in municipal and industrial effluents (Lesage, 1992). Among them, benzene, toluene and ethylbenzene are considered as priority pollutants by the USEPA recently (USEPA, 2014). Toluene is one of the most observed aromatic hydrocarbons in landfill leachate (Meckenstock et al., 2004; Kumarathilaka et al., 2014). Gibbons et al. (1999) have assessed VOC concentrations in old and new MSW dumpsites and found that benzene, toluene, and ethylbenzene concentrations were 40, 212 and 96 $\mu g\ L^{-1}$, respectively, in old landfill leachate, whereas, those figures were 23, 333 and 72 $\mu g\ L^{-1}$, respectively, in new landfill leachate. Additionally, Eganhouse et al., (2001) summarized that wider range of VOC concentrations had occurred in landfill leachate, 1–1630 $\mu g\ L^{-1}$ for benzene, 1–12,300 $\mu g\ L^{-1}$ for toluene and 1–1280 $\mu g\ L^{-1}$ for ethyl benzene. Once these VOCs mixed with groundwater, a high amount of cost is often spent on the remediation.

Individual and combination VOCs effect on ecosystem and aquatic organisms with the groundwater contamination have been reported and exhibited the carcinogenicity effect on human and animal (Peng et al., 2015). Exposure to aromatic hydrocarbons via inhalation, ingestion, and injection would results systemic clinical effects of extensive pulmonary edema and hemorrhage (Reese and Kimbrough, 1993). The acute effects on central nervous system can expect with the VOCs including BTEX at high doses (Shen, 1998). Moreover, low level of chronic exposers are combined with problems in cardio vascular system, kidney, and liver (Liu et al., 2014). Major parcel of the anthropogenic atmospheric VOC source govern by low quality fuel combustion waste management and biomass burning within the urban centers and subsequently, contamination reported in developing countries which is more controversial than reported in developed countries (Chauhan et al., 2014; Sabel and Clark, 1984). In China, average BTEX concentrations in 43 cities were ranged 0.7–10.4, 0.4–11.2, 0.1–2.7, 0.4–15.3 $\mu g\ L^{-1}$, respectively (Barletta et al., 2005). In addition, in Karachi, Pakistan, the reported BTX levels were 16.6, 26.8 and 8.2 $\mu g\ L^{-1}$, respectively (Chauhan et al., 2014).

Although studies on VOCs emission from open dumpsites are limited, it appeared that, VOC emissions related to waste management are accounted as 1–2% from the total anthropogenic VOC sources (Eggertsen and Nelsen, 1958). Benzene reported from landfills in Korea was 32 ± 61.8 $\mu g\ L^{-1}$, considerably higher than the permissible level for drinking water established by World Health Organization (WHO). In contrast, benzene has been reported from Norman landfill in USA was 0–10 $\mu g\ L^{-1}$ (Chauhan et al., 2014), and it is below the WHO permissible level. However, in the absence of proper waste management actions, no systematic monitoring and haphazard practices in developing countries VOCs from open dumpsites would lead to worst case scenario with the health (Vrijheid, 2000). Therefore, due to their severe toxicity, it is a must to take mitigatory measures to reduce their emission to the environment. Hence, monitoring VOCs is indeed a need in order to select the best fortification and alleviation strategies.

Generation of MSW in Sri Lanka is projected to 1.0 kg/capita/day which is 0.8 kg/capita/day at present (Bandara and Hettiaratchi, 2010). On the other hand, the distance between residential areas and landfills are gradually being decreased due to population multiplication and urban boundary expansion. However, a qualitative study on the landfill gas characterization for samples taken from 21 open solid waste dumpsites revealed that potential risk of VOCs within the country (Nagamori et al., 2013). Moreover, some common VOCs including toluene, benzene, dichloromethane and cis-2-dichloromethane are present at those and indirectly indicates the contamination risk of water via leachate. Although VOCs are seem to be highly toxic to biota, no studies have been conducted in quantifying the VOCs present in landfill leachate in Sri Lanka that pose a threat to human and ecosystem health through pollution of river water. With continuous flow of landfill leachate rich in VOCs may deteriorate the sustainability of surface water and also groundwater via infiltration. Therefore, the aim of this study was to assess the quantities of different VOCs present in the landfill leachate from an open dumpsite in Kandy, Sri Lanka during a wet and a dry day in order to observe any seasonal variation.

2. Materials and methods

2.1. Study area and sample collection

Gohagoda open solid waste dumpsite is located in the world heritage city Kandy, Sri Lanka (7°18′ 47.85″N and 80°37′19.02″E). The extent of the landfill is about 2.5 ha. It has been in operation

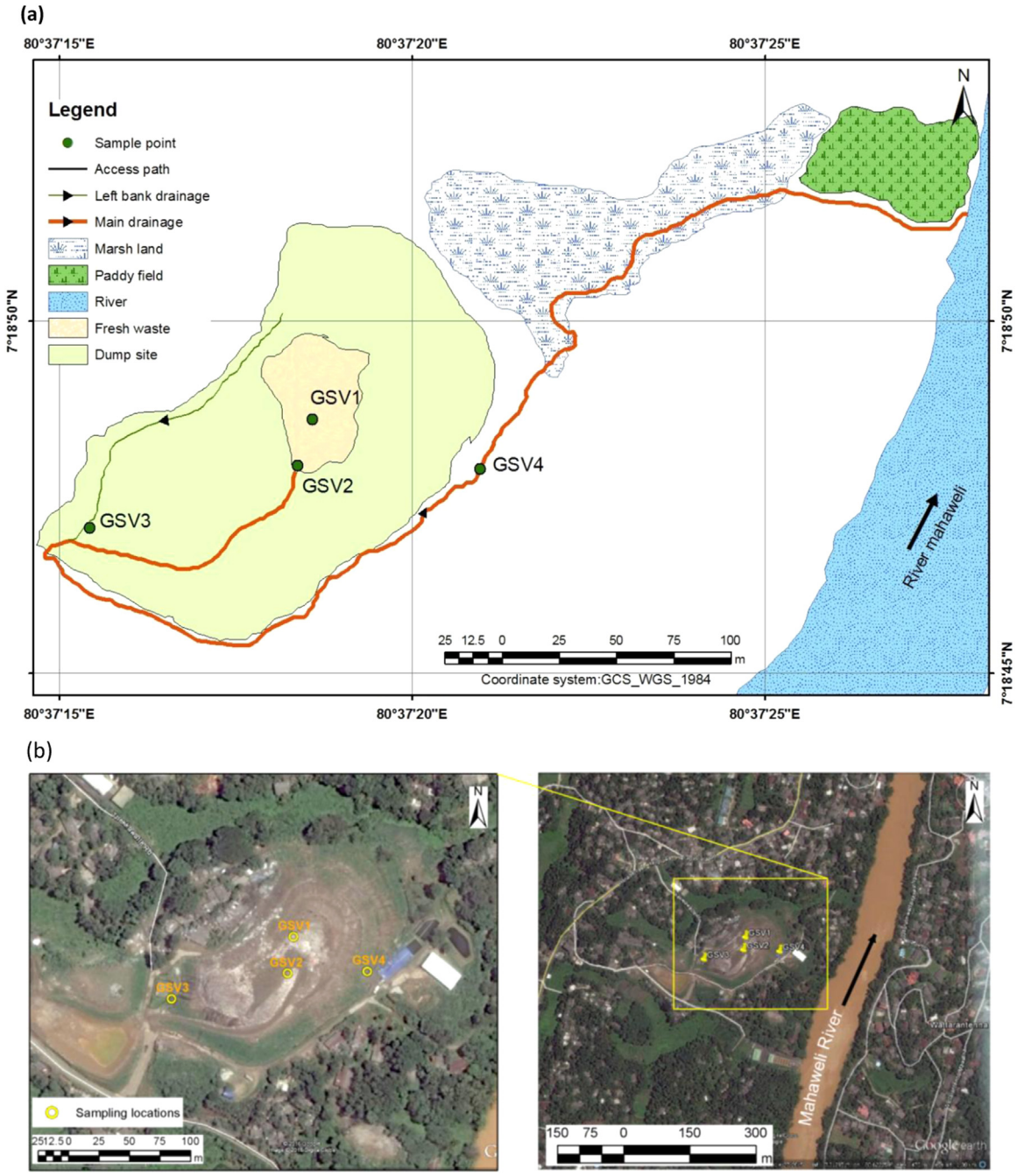

Fig. 1. (a) Schematic diagram of the Gohagoda dumpsite and leachate sampling points (GSV1, GSV2, GSV3 and GSV4) and terrain of the adjacent area (b) and (c).

since the 1960s and receives MSW from the world heritage city, Kandy. At present, it is about 150 tons of MSW consisting waste from households, fish market, slaughter house and non-infectious hospital waste are daily dumped without any pretreatment (Wijesekara et al., 2014). River Mahaweli flows just 100 m distance to the open dumpsite, which is the main water supply source for the Kandy city and suburbs. Leachate from the MSW dumpsite directly flows into the river Mahaweli, the largest and longest river in Sri Lanka, with no treatment. Chemistry of landfill leachate and subsurface flow pattern of the leachate plume have been studied in detail for Gohagoda dumpsite except VOCs (Wijesekara et al., 2015', 2014). Biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD) ranged 1800–3590 and 4800–69,700 mg L^{-1}, respectively, in Gohagoda dumpsite, whereas, the average total organic carbon (TOC) and dissolved organic carbon (DOC) were 37,000 and 28,500 mg L^{-1}, respectively. Additionally, the maximum concentrations of heavy metals including Zn, Cu, Pb, Ni, Cr, and Cd were 0.337, 0.443, 0.356, 0.390, 0.168 and 0.033 mg L^{-1}, respectively (Wijesekara et al., 2014).

All sampling sites (GSV1, GSV2, GSV3, and GSV4) are shown in Fig. 1. GSV1 and GSV2 sampling points locate at the dump, whereas, GSV3 and GSV4 sampling points lay along the leachate draining channel. The field sampling in triplicates was carried out twice based on seasonal sampling, one at the spring (during spring) and autumn (during autumn), 2014 to represent the dry and wet days in the typical tropical climate. The maximum temperature, relative humidity, precipitation and wind velocity were 31.2 °C, 63%, 0 mm and 3.11 km h^{-1}, respectively, on spring 4th. Whereas, the respective values were 28.8 °C, 70%, 19.3 mm and 3.43 km h^{-1} on autumn 15th around Gohagoda. Sampling was

carried out in spring and autumn. During sampling, 1 g of sodium chloride was added to 10 mL of leachate for optimizing resolving peaks in the VOCs and sealed at field conditions (Eichelberger et al., 1989; Restek, 2000). Immediately thereafter, the samples were cooled to 4 °C and transferred to the laboratory for determination of VOCs (Eichelberger et al., 1989).

2.2. Methodology for VOC analysis

VOC analysis was performed using a gas chromatograph (GC-2010 plus, Shimadzu Corporation, Kyoto, Japan) coupled to a mass selective detector (QP-2010 ultra, Shimadzu Corporation, Kyoto, Japan). A headspace sampler (HS-20, Shimadzu Corporation, Kyoto, Japan) was used to partition VOCs into the headspace gas volume and consequently injected into the gas chromatograph. The GC capillary column utilized for all the analysis was Rtx-624 (fused silica column). The detailed operating conditions of this system are presented in Table S1.

The VOC mix analytical standard, prepared in methanol, EPA 524.1 (EPA VOC Mix 2, Sigma-Aldrich Co. LLC., MO, USA) had a concentration of 2000 μg mL^{-1} for each compound. The EPA 524.1 solution contained benzene, toluene, ethylbenzene, *m*/*p*-xylene, styrene, bromobenzene, 1,3,5-trimethylbenzene, 1,2,4-trimethylbenzene, 4-isopropyltoluene, n-butylbenzene, 1,2,4-trichlorobenzene, naphthalene and 1,2,3-trichlorobenzene. The standard stock solution with concentrations of 20 μg mL^{-1} was prepared from the EPA 524.1 solution. The final calibration standard solutions ranged from 1 to 50 μg L^{-1}. The standard curves were in good linear relation (correlation coefficient R^2 = 0.997–0.992). In addition, analytical precision and accuracy in selected method were ranged 0.1–0.01 μg L^{-1}. The VOCs in the landfill leachate were quantified on the basis of their retention times, target and qualifier ions.

3. Results and discussion

Fig. 2 depicts the total ion chromatogram acquired for the mix VOC solution containing 13 standards selected as representative of VOCs in landfill leachate. It appears that the selected temperature program leads to separation of peaks from each other more precisely.

High benzene concentrations were reported in Korea, 32 ± 61.8 μg L^{-1} and in contrast, benzene concentration Norman landfill in the USA was reported as 0–10 μg L^{-1} (Chauhan et al., 2014). Compared to the Norman landfill, benzene concentrations were reported high in the present study and denoted fluctuation based on the rainfall. Concentration ranges of VOCs in the landfill leachate are listed in Table 2. In both sampling periods, landfill leachates contained several compounds. A total number of 9 VOCs were found in spring and autumn. Styrene was detected only during the spring sampling period whereas 1,3,5-trimethylbenzene was found only in autumn sampling period. Bromobenzene, n-butylbenzene, 1,2,4-trichlorobenzene, and 1,2,3-trichlorobenzene were not found in both sampling periods. Benzene and toluene were the most frequently detected VOCs in almost all the sampling locations. A study by Christensen et al. (1994) also reported that benzene and toluene are one of the most commonly observed VOCs in landfill leachate. Benzene concentrations in spring and autumn ranged between 7.59–21.7 and 1.78–2.71 μg L^{-1}, respectively, while toluene concentrations in spring and autumn ranged between 2.71–9.09 and 1.73–20.2 μg L^{-1}, respectively (Fig. 3). The USEPA maximizes a standard limit for benzene as 5 μg L^{-1} in drinking water and groundwater (Liang and Chen, 2010). WHO established the maximum permissible level of benzene, toluene, ethylbenzene, xylene and styrene in drinking water as 10, 70, 300, 500 and 20 μg L^{-1} (WHO, 2008). Hence, it is appeared that the benzene concentration in landfill leachate in spring exceeded the standard limit for the drinking water and groundwater whereas other VOCs, which possess limits established by WHO, does not exceed permissible level in drinking water. Although the concentration per sample is less, with the high amount of leachate flow, the discharge of VOCs may be extremely high compared to the WHO permissible levels. Among the VOCs investigated, 4-Isopropyltoluene had the highest concentration in both periods (38.0 μg L^{-1} in spring and 129 μg L^{-1} in autumn, respectively). Some studies have indicated that 4-Isopropyltoluene become one of the leading odorous components even at trace levels (i.e., 6.36 and 2.89 μg L^{-1} in biogas and leachate, respectively) (Termonia and Termonia, 1999; Davoli et al., 2003). Additionally, 4-Isopropyltoluene concentration in the Gohagoda landfill leachate exceeded the Maine state drinking water guideline value for the 4-Isopropyltoluene, 70 μg L^{-1} (USEPA, 1993). Overall, the sources of VOCs in leachate could be primarily household cleaners, gasoline, lubricating oil, paint, adhesives and synthetic rubber (Sizirici and Tansel, 2010) which were dumped into the Gohagoda MSW dumpsite.

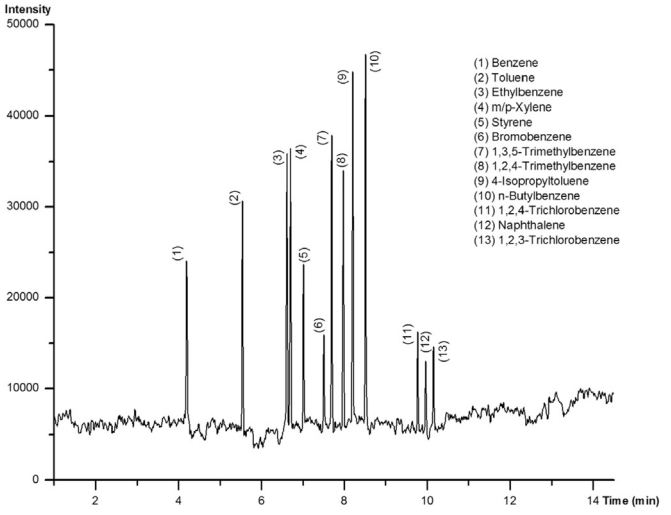

Fig. 2. Total ion chromatogram obtained by headspace 1 μL injection and solution containing 13 VOC standards at the concentration of 10 μg L^{-1}.

The total VOC concentrations quantified and the number of VOCs in a dry (Spring) and a wet (Autumn) month and the sum of different VOCs concentration gives the total VOCs concentration in a particular sampling location (Fig. 4). It appears that samples collected from different locations in spring had higher total VOC concentrations whereas samples collected in autumn had lower levels except GSV1 sampling location. Similarly, a high number of different VOCs was recorded in spring compared to in autumn possibly due to the dilution by rainfall in autumn may have reduced degradation. As it is well known, VOC concentrations in leachate would be highly influenced by the environmental conditions as well as waste handling process (Sizirici and Tansel, 2010). More precisely, various activities that carried out on the landfill site (i.e., unloading and compaction of waste) may affect the level of VOCs in landfill leachate (Chiriac et al., 2007). For this reason, high amount of total VOC concentrations did occur at GSV1 sampling location even during the rainy days. Further, data demonstrate that VOC concentrations and number of different VOCs in landfill leachate decreased as the distance increased from the dumpsite. This observation could be explained by the dispersion of VOCs as soon as they emit into the atmosphere. Thereby, Mahaweli River might not be at potential risk due to the surface flow of landfill leachate in terms of VOCs. Nevertheless, high rainfall in the wet day may transport a greater amount of VOCs even though

Table 2
VOC concentration ranges (μg L^{-1}) at Gohagoda landfill.

	GSV1		GSV2		GSV3		GSV4		WHO permissible limit (μg L^{-1}) (WHO, 2011)
	Spring	Autumn	Spring	Autumn	Spring	Autumn	Spring	Autumn	
Benzene	7.89–21.7	1.78–2.71	7.59–16.1	ND	8.86–18.4	1.83[a]	9.15–15.5	ND	10
Toluene	4.95–9.02	18.6–20.2	5.53–9.09	2.49[a]	3.02–5.20	ND	2.71–4.10	1.73[a]	700
Ethylbenzene	1.30–1.83	3.34–6.36	1.35–1.58	ND	ND	ND	ND	ND	300
m/p-Xylene	1.47–1.58	5.33–6.76	1.45–1.78	ND	ND	ND	ND	ND	500
Styrene	3.25–3.55	ND	ND	ND	ND	ND	ND	ND	20
Bromobenzene	ND	ND	ND	ND	ND	ND	ND	ND	–
1,3,5-Trimethylbenzene	ND	1.57–1.76	ND	ND	ND	ND	ND	ND	–
1,2,4-Trimethylbenzene	2.00–2.26	6.46–7.41	1.47–2.18	ND	ND	ND	ND	ND	70[b]
4-Isopropyltoluene	1.41–14.5	67.4–129	16.4–38.0	ND	ND	ND	ND	ND	–
n-Butylbenzene	ND	ND	ND	ND	ND	ND	ND	ND	–
1,2,4-Trichlorobenzene	ND	ND	ND	ND	ND	ND	ND	ND	70[b]
Naphthalene	5.12[a]	6.03–7.29	4.23–6.91	ND	ND	ND	ND	ND	=
1,2,3-Trichlorobenzene	ND	ND	ND	ND	ND	ND	ND	ND	=

ND – Not detected.

[a] Identified only in one sample.

[b] Maximum concentration limit established by Environmental Protection Agency (EPA) (USEPA, 2011).

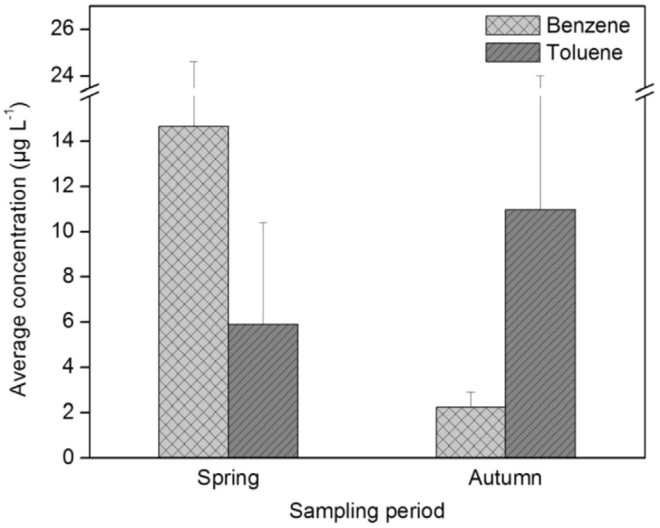

Fig. 3. Variations of average benzene and toluene concentrations in spring and autumn samples.

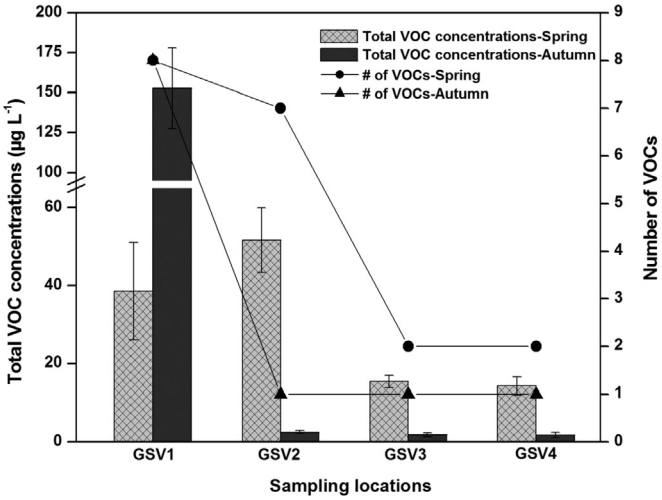

Fig. 4. Variations of total VOC concentrations and number of VOCs in spring and autumn samples.

their individual concentrations are lower. According to the Menikpura et al. (2008),volume of 30,304 m^3 of leachate flows into the Mahaweli River per year. This study revealed that the total average concentration of VOCs is 34.83 μg L^{-1}. In this sense, desired loading of the VOC content is 2.89 g day^{-1} on average basis. In addition, the subsurface flow of landfill leachate consisting VOCs may possibly be contaminating Mahaweli River since a resistivity survey revealed that subsurface flow of Gohagoda leachate flows directly to Mahaweli River (Wijesekara et al., 2015).

4. Conclusions

It is the very first study on the quantitative assessment of VOC concentrations in the leachate, which flows from an open dumpsite of MSW in Sri Lanka. Overall, the dumpsite had higher total VOC concentrations in spring compared in autumn except GSV1 sampling point (152.78 μg L^{-1} in autumn). It was found that total VOC concentrations and the number of VOCs detected were high in the middle of the dumpsite, where fresh municipal waste is dumped, compared to the locations lying along the leachate drainage channel. It may be due to the volatilization of VOCs with the distance of the channel from the dumpsite. Although the total concentrations of VOCs exhibited a reduction with the distance from the dumpsite, their adverse effects are not negligible due to the enormous quantity of leachate flows downstream. At the same time, due to the accelerated urbanization around Gohagoda area, the dispersion of VOCs into the air may influence the quality of life in nearby residents. It is suggested that although the concentrations were less per sample collected with the large flow of leachate due to the high amount of rainfall, VOC discharge far exceeds the permissible levels. Furthermore, this preliminary quantification of VOCs in Gohagoda landfill leachate may assist in the development of efficient remediation strategies to remove excessive amounts of VOCs from landfill leachate.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.gsd.2016.04.001.

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