Contents lists available at ScienceDirect

Journal of Hazardous Materials

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



Hazardous organic compounds in urban municipal solid waste from a developing country

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ARTICLE INFO

Article history: Received 23 January 2007 Received in revised form 13 December 2007 Accepted 29 February 2008 Available online 8 March 2008

Keywords: Hazardous organics MSW TCLP Cresol Phthalate

ABSTRACT

Fresh and partially decomposed municipal solid waste (MSW) collected from three places in Chennai city, viz., a residential collection point and two dumping grounds (Kodungaiyur and Perungudi) were screened for hazardous organic pollutants. Toxicity Characteristics Leaching Procedure (TCLP) using a Zero Headspace Extractor (ZHE) followed by further extraction by solvent separation using *n*-hexane containing 15% di-ethyl ether was performed and the organic extract obtained was qualitatively screened by GC-MS. 28 different types of higher alkanes and their derivatives, 7 types of C6–C8 fatty acids and their esters, 7 different phenolic compounds including alkylated phenols and degradation products and 5 phthalate compounds occurred in a majority of the analysed samples. 17 other organic compounds such as carboxylic acids, chloroform, phosphate, pharmaceutical chemicals etc. were also detected. Among these compounds, phenolics and phthalates are highly hazardous in nature and occurred in relatively higher concentrations. Hazardous compounds like *p*-cresol, di-butyl, mono butyl and di-ethyl pthalates were found in concentrations more than 200 mg/kg in MSW.

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1. Introduction

A variety of hazardous organic compounds occur in Municipal solid waste (MSW). Studies from developed countries like Denmark, Sweden, Germany, the United Kingdom and the United States have confirmed the presence of hazardous organic contaminants such as phthalate compounds, phenolics, pesticides, volatile compounds such as benzene, toluene, ethylene and xylene (BTEX), polyaromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) both in the municipal landfill leachates and gases since the late eighties [1–11].

Schrab et al. [12] and Kjeldsen et al. [7] found that, even without landfill co-disposal, leachates from MSW are very similar in composition to those from mixed or hazardous landfills. Table 1 gives a categorical listing of the different organic compounds in municipal landfill in the different regions of the developed world as reported in original research papers and reviews, so far. A scarcity of literature with respect to similar studies in the developing world could be realized and this necessitated the present study. Oman [29] first indicated that organic compounds are primarily present at μ g/L concentrations in landfill leachates and that, in exceptional cases, leachates have been sampled with relatively high mg/L concentrations of organic compounds as well.

These compounds constitute only a minor fraction of the organic portion of MSW. Nevertheless, much concern is associated with these groups of compounds as many of these are highly toxic and refractory in nature posing potential health and environmental risk [30]. The present study has attempted to address this concern in the context of MSW generated in a typical urban scenario in a developing country. The primary objective was to develop the profile of specific organic compounds and to quantify the predominant hazardous pollutants possible in the worst-case of leaching from the MSW. For this purpose, both fresh MSW and MSW mined from an open dumpsite in a metropolitan city, Chennai, in South India, were extracted using toxicity characteristics leaching procedure and screened for hazardous organic compounds.

2. Materials and methods

Chennai, the fourth biggest metropoliton city in the country has the highest per-capita MSW generation rate of 0.45 kg/day. Two open dumping sites are in operation since 1987 in the city, one at Kodungaiyur and the other at Perungudi, to dump the municipal solid wastes collected from south and north zones of Chennai Corporation, respectively. These sites are located at about 10 km from the central part of Chennai city. Both sites fall in seismic zone II of India. An Integrated approach to assess the risk posed by these two dumps revealed that they pose moderate environmental risks [31]

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^{0304-3894/\$ -} see front matter © 2008 Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2008.02.111

Table 1

A listing of organic compounds in municipal landfills identified in previous studies

S.No	Category	Organic compounds identified in municipal landfill emissions	References
1 2	Aromatic hydrocarbons Phthalate compounds	Alkylnaphthalenes, fluoranthene, tetrachloroethylene, benzene, xylene, toluenes and their alkyl derivatives Phthalic acid, di-ethyl phthalate and mono ethyl phthalate, 2-ethyl hexyl, butyl, ethyl, methyl and nonyl esters. terephthalic acid, isoterephthalic acid	[13,14,10] [6,7,10,11]
3 4	Phenolics Pesticides	Phenol, Cresols, nonylphenols, isomers of methylphenol and ethylphenol, chlorocresol isomer, Bisphenol A Ametryn, atrazine, bentazon, chlorpropham, dichlobenil, chloridazone, glyphosate, hexazinon, isoproturon, propoxuron, 4-chlorophenoxy propionic acid and 2,6-dichloro phenoxy propionic acid, Mecoprop, dichloroprop, mechloroprop, hydroxyl simazine, hydroxyl atrazine, desethylatrazine, disisopropylatrazine, <i>N</i> , <i>N</i> -diethyltoluamide, bentazon, MCPA	[15–19,6,20,10] [1,21,22,2,5,7,8,10]
5	Adsorbable Organic Halides	Chlorinated volatile compounds	[15-17,6]
6	VOCs	Dioxanes, tetrachloroethylene and other volatile organic carbons, chlorofluorocarbons, methyl- <i>tert</i> -butyl-ether (MTBE)	[16,15,1,23,17,24,7]
7	Organotin	Mainly monobutyltin	[25]
8	Others	Caffeine, propyphenazone, sulphonamides, barbiturates, phenazone, pentobarbital, Propyphenazone, ibuprofen and clofibric acid, C1–C4 alkylated pyrazines, cyclopentapyrazines, pyridines, short chain C2–C7 alkanoic acids, the corresponding hydroxylated compounds and branched C4–C9 alkanoic acids and C4–C9 dicarboxylic acids, terpenoics, terpeniol, methylnopinone and 1,8-cineole, skatole, tris (2-chloro-ethyl) phosphate, bi-cyclo compounds like camphor and fenchone, nicotine, benzthiazol, cotinine, benzthiazolones	[26–28,6,11]

and hence these two sites were included in the sampling program apart from a nearby waste collection station.

2.1. Site of sampling

Fresh residential MSW was collected from a waste collection point and partially degraded MSW was mined from the two major dumping grounds in Chennai, *viz.*, Kodungaiyur Dumping Ground (KDG) and Perungudi Dumping Ground (PDG).

KDG spreads to over 160 ha in a marshy land adjacent to Kodungayur sewage treatment plant on the southern regions of flood prone alluvial low lands of Korattalaiyar river. The current dumping rate is about 1800 t/day [32]. It is located 2.5 km east of the Kodungaiyur village and lies at $13^{\circ}07'37.6''$ N and $80^{\circ}16'48''$ E.

PDG is a low-lying site with a total area of about 250 ha in which about 22 ha is used for open dumping. The current dumping rate is about 2200 t/day [32]. It lies at 12°57′13.5″N and 80°14′5.8″E. PDG is located 1.2 km south of the Perungudi village and is close to the sea level. It is poorly drained and consists of an extensive area of marshy land which is permanently wet and seasonally inundated.

The average depth of waste filling at both KDG and PDG is about 3–4 m. Disposal of mixed hazardous waste such as health care waste and industrial sludges has been occasionally reported in both the dumping grounds. Open burning and rag-picking by informal sectors and stray animals are very common.

Random grab samples were collected at five different locations from each dumping ground. Both surface and depth sampling were carried out. Samples were collected manually from the surface of the open dumps, an auger was used to collect samples from a depth of 1 m and a backhoe excavator was used to collect bulk samples from over 2–3 m depth. About 5 kg of MSW was collected in clean, labeled, airtight polythene bags in each location and transported to the laboratory for further investigation.

Leachates found stagnant at two locations in each dumping ground and ground water samples from two borewells adjacent to each dumping ground were also collected. These liquid samples were collected in sterilised 1 L glass bottles to detect the presence of leachable organic compounds. Primary physicochemical characteristics of the MSW and leachate samples were determined according to the standard APHA methods [33].

2.2. Leaching procedure, extraction and analysis

The MSW samples collected were shade dried, ground and reduced to particle size less than 9.5 mm. Toxicity Characteristics Leaching Procedure (TCLP) was done to extract the organic compounds from the MSW using a Zero Headspace Extractor (TCLP-ZHE. YT30 090 HW, Millipore, India) according to the standard procedure [34]. Upon addition of 3.5 mL 1N HCl to 5.0 g of MSW (to which 96.5 mL of reagent water had been added), heating at 50 °C for 10 min and cooling, the pH of all the MSW samples dropped below 5.00. Hence, the extraction fluid selected was 0.1 N acetic acid containing 64.3 mL of 1N NaOH/L (pH 4.93 ± 0.05). A known amount of MSW was weighed out such that the ratio of the solid phase (TS of the MSW) to the extraction fluid equaled 1:20 and was pressurized in a ZHE vessel not exceeding a maximum pressure of 50 psi. After pressurization, 500 mL of the extraction fluid was dispensed into the ZHE. The contents in the vessel were agitated for 18 h at 30 rpm. After the extraction period of 18 h, the TCLP extract of MSW was pressurized, collected and further extracted with 15% di-ethyl ether in *n*-hexane for the analysis of organic compounds by Mass Spectrometer (QP-2010, Shimadzu, Japan) linked to a Gas Chromatograph (GC-2010, Shimadzu, Japan), i.e., GC-MS.

The column used for detecting the organic compounds by GC-MS was RTX 624 fused silica capillary column of 30 m length, 0.32 mm pore size and 1.8 μ m film thickness. The injection was carried out at 40 °C, 15 min hold, then programmed to 10 °C/min to 240 °C. The split ratio was set as 1:10. Helium gas was used as the carrier at a flow rate of 1.34 mL/min. An injection volume of 1 μ L was used for analysis and the total run time was 30 min.

3. Results and discussions

The fresh MSW contained higher moisture (average of 50%) and organic contents (VS: 40–50% and organic carbon: 20–30%) than the MSW mined from the dumping grounds (moisture: 13–30%, VS: 9–35% and organic carbon: 4–10%). A diverse range of organic compounds could be expected from such organically rich MSW with heterogenous and complex composition. Leachates collected from the dumping grounds exhibited a wide range of organic content as indicated by the COD (88–15,200 mg/L), DOC (38–5300 mg/L), BOD (48–7557 mg/L) and VFA (90–1080 mg/L). The ground water samples had a low conductivity ranging from 1 to 4 mS/cm as compared to leachates (10–41.5 mS/cm). The organic load in the water samples was very low (COD, DOC and BOD well below 100 mg/L and VFA below detectable levels).

3.1. Occurrence of organic compounds

Table 2 gives an exhaustive listing of organic compounds identified by GC-MS screening in the waste and leachate samples. Higher alkanes and their derivatives, fatty acids and their esters, phe-

 Table 2

 List of organic compounds identified by GC-MS screening in the present study

S.No	Organic compound	TCLP extract		Leachate from	Remarks on health or environmental hazards		
		Fresh MSW	Mined MSW	dumpsite			
I	Higher alkanes and their derivatives						
1	Dodecane		+		Include a wide array of petroleum or crude oil components		
2	Tridecane	+	+		Cycloparaffins and isoparaffins (isoprenoids) are more		
2	Pontadocano		1		persistent than paraffins.		
2	Pentauecane		т		17 and 23) are probable depressants to the central nervous		
					system (CNS) and irritants when present in high		
					on the viscosity of the compound and may cause chemical		
					pneumonitis. Narcosis is one of the major toxic effects [36].		
					Trichloroethane is a fumigant and has proven narcotic properties, is a local irritant to eves, nose and lungs, may		
					be injurious to liver and kidney [35].		
4 5	Hexadecane n-Nonane	(+)	+ +				
6	2-Methylpentane		(+)	+			
7 8	3-Methylpentane Methylcyclopentane		(+) (+)	+			
9	2,2-Dimethyl butane		+				
10 11	2,2,3-Trimethyl pentane		+ +				
12	3,4-Dimethyl undecane		+				
13 14	2,3-Dimethyl hexane	+	+				
14	3,8-Dimethyl undecane		+	+			
16 17	4,7-Dimethyl undecane		+				
17	Trichloroethane		(+) (+)				
19	Cyclohexane,1,5,1,5 di-isopropyl-2-3-dime		+				
20 21	n-Nonadecane	+ +	+				
22	n-Heptadecane	+	+				
23 24	n-ietradecane 2,2,4,6-Tetramethyl heptene		+	+			
25	3,7-Dimethyl nonane			(+)			
26 27	5-Methyl-5-propyl nonane 6-Ethyl-2 methyl decane		+	+			
28	2,6,11-Trimethyl dodecane		+				
II	Fatty acids and their esters						
1	Heptanoic acid Hexanoic acid	(+)	+ (+)		Short-chain fatty acids are probable irritants [35].		
3	Octanoic acid		+				
4	Heptanoic acid propyl esters Heptanoic acids Ethyl esters		+ +				
6	Hexanoic acids butyl esters		+				
7	Hexanoic acid propyl esters		+				
III 1	Phenolics and their degradation products	(+)			Crocole have corrective action on the skin and mucous		
1	p-cressi	(')			membranes producing severe chemical burns and		
					dermatitis. Absorption may result in damage to kidney,		
					cresol [37].		
2	o-Cresol	+			2. Ethyl sharel may load to some disarders [25]		
3 4	3-Ethyl phenol Hydroquinone or 1,4-benzenediol		+		Hydroquinone can be fatal by ingestion, cause dermatitis		
					on skin contact, keratitis and dicolouration of conjunctiva		
5	Acetophenone	+			Acetophenone is narcotic in high concentrations [35]		
6	p-Benzoquinone	+			p-Benzoquinone causes damage to skin and mucous		
					disturbances to vision on eve contact [35]		
7	4-Methyl-2,6-di-tert-butyl phenol		+	+			
IV	Phthalates and their degradation products						
1 2	Di-butyl phthalate Butyl phthalate	(+) +	+ +		Phthalates are hazardous and persistent. Result in reproductive difficulties liver problems		
-					increased risk of cancer and can be estrogenic with a		
3	Di-ethylphthalate	+	+	+	cumulative action.[38,39]		
4	Di-iso butyl phthalate	(+)	(+)				
5	Phthalic acid, butyl 8 methylnonyl ester		+				

Table 2 (Continued)

S.No	Organic compound	TCLP extract		Leachate from	Remarks on health or environmental hazards	
		Fresh MSW	Mined MSW	dumpsite		
v	Others					
1	Cyclohexanoic acid		+		Tri butyl phosphate causes stimulation of CNS. Benzoic acid and fumaric acid are mild irritants. Chloroform causes toxic effects on liver and heart and is a suspected carcinogen. p-Cymene may lead to narcosis, anemia, leucopenia and enlarged liver [35].	
2	Tributyl phosphate		+			
3	Lidocaine		+			
4	Benzoic acid	+	+			
5	Benzoic acid, butyl ester	+				
6	Carbonic acid, phenyl propyl ester		+			
7	2-butenedioic acid (fumaric acid)		+			
8	Benzene sulfonic acid, 4-methylbutylester		+			
9	Chloroform		(+)	+		
10	Octa-sulfur		+	+		
11	N,N,N,N'-Tetra ethylenediamine			+		
12	p-Cymene			+		
13	Acetamide, 2-(diethylamino)-N-(2,6-dii)			+		
14	Hydroperoxide,1-ethylbutyl	+				
15	Hydroperoxide, 1-methylpentyl	+				
16	Heneiocosane	+				
17	2,4,4,6-Teramethyl-2-heptene			+		

Note: '+' denotes presence of the compound in the TCLP extract or leachate; '(+)' denotes occcurence in relatively higher no. of samples than others.

nolic and phthalate compounds and their degradation products occurred in almost all the samples. Phthalate compounds were more common in both fresh MSW and MSW mined from the dumping grounds. Phenolic compounds were abundant in the fresh MSW.

The hazardous organic compounds are primarily leached out from the hazardous materials disposed into the dumpsite. They might also be the by-products and end-products of various biochemical processes occurring naturally within the dump or the result of open burning activity in the dumping grounds. The hazardous chemicals could have originated from hazardous domestic wastes or illegitimately dumped hazardous industrial sludges and solid wastes. The possible specific sources of residential origin include pharmaceuticals, soaps and cosmetics, plastics, resins, textile, leather preservatives, lacquers, varnishes, solvents, etc. [2].

Higher alkanes like nonanes and decanes are components of fuel oil, lubricating oil and paraffin wax. The abundant occurrence of fatty acids and their esters, *i.e.*, C2–C7 alkanoic acids has formerly been described as a result of anaerobic fermentation in addition to the short chain alkanoic acids, the corresponding hydroxylated compounds as well as branched C4–C9 alkanoic acids and C4–C9 dicarboxylic acids. Ethyl, butyl and propyl esters of alkanoic acids and cyclo alkanoic acids are derived mainly from the microbial degradation of lignin [6]. Reinhart and Pohland [40] have reported that the fatty acids can be formed due to decomposition of lipids (2 to 12 C fatty acids), proteins and carbohydrates. Short-chain fatty acids make up 49% of the dissolved organic carbon [41] and are bound to occur in anaerobically degrading MSW.

Phenolic compounds have both anthropogenic (petroleum refining, gasification, preparation of solvents, paints and varnishes) and natural (canopy leachates, leaf litter, root exudates and microbial metabolism) origins [42]. Phenolic compounds such as cresols, may be degradative by-products of lignin.

Phthalates are highly persistent compounds. These organic plasticisers are added to Poly Vinyl Chloride (PVC) products to improve their processing properties [25]. Certain low-molecular weight phthalates are used in personal health care products. The occurrence of phthalic acids and its hydrogenation products have been reported to be due to transformation process of phthalates in waste deposits via. hydrogenation and hydrolysis [6]. Mono-esters of phthalic acids originate from degradation of di-esters of phthalic acids [10]. A chlorinated compound, trichloromethane, occurred in a quite a few number of dumpsite samples. Such chlorinated aliphatic hydrocarbons have been previously found in a relatively large number of landfills [7]. These halogenated compounds are relatively more persistent in the environment and therefore much concern is attached to them than others. They are used in dry cleaning and as solvents [43].

An alkyl ester of phosphate plasticizer, tributyl phosphate, was also detected. The presence of octasulfur in KDG MSW shows the oxidation of sulfide compounds and creosite. Acetamide compounds and lidocaine, which is a local anaesthetic and antiarrhythmic drug, indicate the disposal of pharmaceutical drugs and chemicals in the municipal stream.

Carboxylic acids and their alkyl esters were also detected in a few samples. Benzoic acids and their butyl esters may be the degradation product of alylbenzenes [15] and may also be intermediates of the methanogenic degradation of ferulic acid, a lignin compound [44].

3.2. Quantification of selected hazardous organic compounds

A selected few of the predominant organic compounds which have proven hazardous nature were quantified. Table 3 lists the concentrations at which such predominant hazardous organic compounds occurred in the screened samples. Typical GC-MS scans of the TCLP extract and the open leachate are depicted in Figs. 1 and 2, respectively.

3.2.1. Phthalate compounds

Among the phthalate compounds, di-butyl phthalate (DBP) occurred upto even 6932 mg/kg in fresh MSW. Other phthalates like di-ethyl phthalate (DEP) and di-isobutyl phthalate (DIBP) were also found in considerably higher concentrations in fresh (1174.70 and 1389.23–1426.43 mg/kg, respectively) and mined MSW (403.82–1105.38 and 1312 mg/kg, respectively). Mono-butyl phthalate (MBP), which might be a by-product of degradation of the higher phthalate compounds, was detected only in a single dump-site sample indicating the persistence of phthalates even years after dumping.

Gray et al. [39] revealed that male reproductive development is acutely sensitive to some phthalates like DBP and di-ethyl

Table 3

Concentrations of selected hazardous organic compounds in MSW, leachates and ground water

Hazardous organic compound*	Fresh MSW (KCS)	Mined MSW (KDG and PDG)	Dumpsite leachate	Ground water
Phthalates				
Di-butyl phthalate	196.20-6932.44	264.85-1122.94	BDL	BDL
Di-ethyl phthalate	BDL-1174.70	403.82-1105.38	56.8-495.3	BDL
Di-iso butyl phthalate	1389.23-1426.43	BDL-1312	BDL	BDL-0.012
Mono butyl phthalate	BDL	BDL-325	BDL	BDL
Phenolics				
p-Cresol	440.22-1899.5	BDL-258.72	583.9	BDL
o-Cresol	BDL	BDL-243.26	BDL	BDL
Higher alkanes				
Pentadecane	BDL	BDL-1170.26	BDL	BDL
Hexadecane	482.02-805.76	218.04-752.31	BDL	BDL
Heptadecane	BDL-104.18	BDL-966.33	BDL	BDL
Nonadecane	BDL-657.18	BDL-400.53	BDL	BDL

Note: * Concentrations of the compounds are expressed in mg/kg for MSW and in µg/L for leachate and ground water. BDL: below detectable levels of 0.001 mg/kg for MSW and 0.001 µg/L for leachate and ground water.



Fig. 1. A typical GC-MS scan of TCLP extract of MSW showing the major organic compounds.

hexyl phthalate (DEHP). Van Wezel et al. [45] have established an environmental risk limit (ERL) of 0.7 and 1 mg/kg of fresh weight for DBP and DEHP, respectively, for fresh soils and sediments with atleast 10% organic matter. Concentrations exceeding the above ERLs would cause endocrine disruptive effects. They have also indicated that in Netherlands, DEHP concentration is 3–20 times higher than the ERL. In the present study, the concentration of DBP was remarkably higher than the ERL. This is due to the occurrence of flexible PVC plastic waste as a major component of unsegregated MSW and discarding of a considerable amount of personal health-care products in the municipal stream.

Leachate from the dumping grounds contained only DEP at a concentration ranging from 56.8 to 495.3μ g/L. Screening of the borewell water in the area adjoining the dumping ground revealed the presence of only two compounds, *viz.*, DIBP and 3,7-dimethylnonane in the ground water. As expected, the phthalate compound DIBP has leached into the underlying ground water as it was more predominant in MSW among all the other phthalate compounds. Van Wezel et al. [45] have established ERLs of



Fig. 2. A typical GC-MS scan of open leachate from dumpsite showing the major organic compounds.

10 and $0.19 \mu g/L$ for DBP and DEHP in water. The USEPA [46] has established drinking water equivalent levels (*i.e.*, a lifetime exposure concentration protective of adverse, non-cancer health effects assuming that all the contaminant is from drinking water) of DBP and DEP as 4 and 30 mg/L in drinking water. The concentration of DIBP (0.012 $\mu g/L$) was below these safety thresholds for drinking water. Nevertheless, leaching and potential contamination of ground water with other phthalate compounds in the near future is anticipated.

3.2.2. Phenolics

Among the phenolic compounds, p-cresol occurred frequently in the fresh MSW at a concentration ranging 440.22–1899.5 mg/kg. In the dumpsite, o-cresol and p-cresol occurred only in a single sample. The phenolics concentrations were well below the hazardous limit of 5 g/kg established for phenolic compounds in Schedule 2 of Hazardous waste (Management and Handling) Rules [47]. p-Cresol occurred at 583.8 μ g/L in the dumpsite leachate. This concentration is below the phenol concentrations (1 mg/L) allowed for safe disposal of leachate in water bodies [48]. An alkylated complex phenolic, *viz.*, 4-methyl-2,6-di-*tert*-butyl phenol was also found in the dumpsite leachate. The present finding agrees with the previous reports that cresols (used in wood preservatives, drugs and detergent manufacture) are the primary phenolic compounds occurring in high concentrations in MSW [35,6,10].

3.2.3. Higher alkanes

Hexadecane was detected at high concentrations in both fresh MSW (482.02–805.76 mg/kg) and MSW mined from the dumpsites (218.04–752.31 mg/kg). Other higher alkanes like hepta-, octaand nona- decanes were also detected each in a single sample of fresh and mined waste with pentadecane and heptadecane at particularly higher concentrations (1170.26 and 966.33 mg/kg) in the dumpsite samples.

Approaches to tackle the problem of these hazardous compounds in MSW may include minimization of hazardous contents at source by judicious use and recycling or source segregation of the generated waste for management along with similar hazardous wastes of industrial origin.

4. Conclusion

Higher alkanes, fatty acids, phenolics, phthalates and their respective degradation products were predominant in the TCLP extracts of both fresh and mined MSW. Fatty acids are nonhazardous by-products of anaerobic digestion of MSW while a few higher alkanes, phenolics and phthalates are potentially hazardous compounds with proven adverse impact on both human health and environment.

Among the phenolics, *p*-cresol was found to occur abundantly in fresh MSW (1899 mg/kg) than mined MSW (258 mg/kg) indicating phenolic degradation when disposed with MSW over a longer time. Phthalates were found in both fresh and mined MSW at comparable concentrations indicating the persistent nature of these pollutants. The leachate samples were screened positive for two hazardous compounds, *viz.*, DEP (upto 495 μ g/L) and p-cresol (583 μ g/L). Only one phthalate compound, di-iso butyl phthalate, was detected in the ground water at a concentration of 0.012 μ g/L.

Acknowledgements

The authors wish to acknowledge the financial support provided by the Swedish International Cooperation Development Agency (SIDA) for the realization of this study. The cooperation extended by the Corporation of Chennai (CoC) and the Asian Institute of Technology (AIT) is thankfully acknowledged.

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