

EVALUATING FUGACITY MODEL FOR ORGANIC CONTAMINANTS IN WASTE DEPOSITED IN KHULNA LANDFILL

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ABSTRACT

This study was undertaken to assess the behaviour of some toxic organic contaminants like Atrazine, Trichloroethylene, and Benzene etc. in different landfill components such as landfill gas (LFG), leachate and waste due to emission from a selected waste disposal site at old Rajbandh, Khulna, Bangladesh. Level III Fugacity model was implemented on the selected evaluative environment and Monte Carlo simulation was used to account the variability and uncertainty of the model inputs as well as to observe its effect on the model outputs. It was found that Trichloroethylene had the highest concentration in waste compartment with a magnitude of $2.0E-02 \text{ mol/m}^3$ and most of the mass (52%) was accumulated in waste compartment. It was found that *m*-Xylene was the highly persistent organic contaminant as it spends the highest amount of time in the modelled landfill environment. Reaction was the main removal mechanism for Trichloroethylene as about 79% of the total amount was removed by oxidation and hydrolysis reaction. It is essential to know the behaviour of potential harmful contaminants for assessing human health hazards from landfill site. The outcome of level III Fugacity model like mass, concentrations, etc. of organic contaminants generated from a selected landfill will further be helpful for evaluating health hazards from landfill site at Khulna.

Keywords: Fugacity model; Monte Carlo Simulation; Organic contaminants; Advection; Reaction; Intermedia transport.

1. INTRODUCTION

Landfill is a unit operation system for municipal solid waste (MSW) disposal and it must be designed to protect the environmental receptors such as human, water, landfill gas (LFG), soil, etc. from contaminants which may be present in the waste stream (Visvanathan *et al.*, 1999). The deposited MSW in landfill then decomposes and various types of toxic compounds such as Atrazine, Trichloroethylene, Benzene, etc. generates from LFG and leachate (Prince *et al.*, 2018; Yin *et al.*, 2019). These contaminants enter the atmosphere and pollute the main components of environment such as atmosphere, lithosphere (land) and hydrosphere (water) (Shenbagarani, 2013; Olafisoye *et al.*, 2013). The hypothesis is that the landfills can be a source of environmental pollution and risk. The principal sources of emissions from landfill sites are as follows: the waste materials as they are brought onto site; emissions from transport; MSW blown by the wind; dust generated from landfill surface; LFG generated; leachate produced (Magdalena, 2019). Leachate and LFG are the two crucial and principal outputs of landfill. These leachate and LFG are the dominant components for the environmental impacts as well as public health effects as they contain harmful toxic chemicals having carcinogens and non-carcinogens behaviour (Rafizul and Pangkaj, 2019; Marinella *et al.*, 2014). Several contaminants found in the landfill tend to bio accumulate and produce toxic effect at low concentrations. Contaminants of potential concern commonly found in LFG include xylene, toluene, hexane, benzene, trichloroethylene, chloroform and so on (US.EPA, 1997). Organic compounds that are hazardous at low concentrations may be present, e.g. pesticides (atrazine, simazine), AOX (adsorbable organic halogens) compounds, etc. Most are manmade but some may be formed within landfill (US.EPA, 1997).

Many types of contaminant fate models are available in the literature. They fall into two distinctive types: those that follow a multimedia fugacity approach and those that follow a concentration-based analysis (Chao *et al.*, 2019). Models that follow the fugacity approach use fugacity instead of the conventional concentration for the quantification of all contaminants of concern in the system. The use of such an approach simplifies the modeler's understanding of the path of contaminant fluxes from one phase to the next (e.g., soil to water, water to air, and so forth) and reduces the number of constants used in the model (Kilic, 2008). Therefore, it has become important to know the possible concentration, mass and percentage of these chemicals in different waste components like LFG, leachate and waste (soil) to extract the behaviour profile of these contaminants in the environment through models. Fugacity is the tendency of a chemical to escape from a medium. It is the pressure that the chemical exerts on a medium and the chemical potential of a substance in that medium (Gobas and MacLean, 2003). In addition, Fugacity is the chemical equilibrium criterion between different phases and can be used as a tool for predicting the behavior of contaminants in different landfill phases. It simplifies the understanding of an equilibrium between different phases as they reach a common Fugacity while attaining

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different concentrations in different evaluative media. As chemical seek equal Fugacity between different phases, Fugacity gradient becomes the driving force for a chemical to move from one media to another while equilibrium is not reached between them. Chemical fugacity is in equilibrium within different phases of each compartment but is in non-equilibrium among the compartment themselves (Linyu Xu *et al.*, 2015). In general, Fugacity is a thermodynamic quantity used as an equilibrium criterion with units of pressure (Pa) (Kilic, 2008). Fugacity model is a great tool for predicting the behaviour of contaminants which are subjected to steady state partitioning, advection, reaction, intermedia transport etc in an evaluative environment. This model generates consistent behaviour profiles which may be useful for predicting the behaviour of contaminants for which no environmental observations exist (Yan *et al.*, 2013). In addition, multimedia Fugacity models can be categorized into four levels in the literature: Levels I, II, III, and IV (Mackay, 2001). In Level I Fugacity models, steady-state and equilibrium conditions are assumed between environmental compartments which are so unrealistic in comparison with the real environment. In Level II fugacity models, chemical transformations and advection are considered with the assumption of steady-state and equilibrium conditions, which is also rarely the case in the real environment. Level III models assume steady-state, but non-equilibrium conditions between environmental compartments. Level IV models consider non-equilibrium conditions between environmental compartments under unsteady-state conditions. Level IV approaches are the most realistic. However, Level IV approaches are seldom applied due to their complexity and data requirements.

In this study, an attempt has been made to find the behaviour of four priority contaminants like atrazine, trichloroethylene, m-xylene and benzene that are likely to be generated from landfill on the basis of Level III Fugacity model. They are considered to be formed in the landfill evaluative environment as a result of decomposition of agricultural, homemade and industrial waste dumped into the landfill. The behaviour mean concentration, mass distribution, fugacity, rate of reaction, rate of advection, rate of intermedia transport, residence time etc. of selected contaminants which releases from an open disposal site at old Rajbandh, Khulna. This study sought to build a representative landfill environment derived from fictitious but realistic properties such as composition, volume and temperature as well as apply the Level III Fugacity model. The objectives of this study were to (i) characterize the fate of these contaminants in the evaluative environment based on level III Fugacity model and (ii) perform Monte Carlo simulation using @RISK 7.6 (Palisade, 2019) to validate the obtained behaviour profiles of these contaminants and find the most sensitive parameters. The outcome of level III Fugacity model like mass, concentrations, etc. of organic contaminants generated from a selected landfill will further be helpful for evaluating health hazards from landfill site at Khulna.

2. FUNDAMENTAL THEOREM OF THE STUDY

Assessment of chemical fate requires some modelling because human mind is incapable of assimilating and processing the various disparate quantities such as vapor pressure, octanol water partition coefficients, reaction rate constants and transfer coefficients which combine to determine the chemical's behaviour (Mackay *et al.*, 1985). The Level III Fugacity model adopted here comprises three environmental compartments: LFG, leachate and waste. The model in this study was generated in the form of a MS Excel program. Areas, depths and volumes are user specified from which the volume of each compartment was derived. Other input parameters include advection inflow rate (G), fraction organic carbon and emission rate of the contaminant in each of the compartment. With regards to the chemical input data, molecular weight, density, aqueous solubility (C_s), temperature (T), vapor pressure (P_s), first order reaction rate constant (K), octanol water partition coefficients (K_{ow}), mass transfer coefficients (K_{ij}), molecular diffusivity etc. need to be specified. Using the input parameters, Henry's Law Constant (H) is determined using the following Equation 1,

$$H = \frac{P_s}{C_s} \quad (1)$$

Where, H is the Henry's Law Constant ($\text{Pa}^3\text{m}/\text{mol}$), P_s is Vapor pressure (Pa) and C_s is Aqueous solubility (mol/m^3). Biodegradation, hydrolysis, photolysis, and oxidation reaction were considered in each of the compartment for the decay or removal of the contaminant from the evaluative environment. First order kinetic expression was considered for all these reactions. Though there are many situations in which the real reaction rate is not a first-order reaction. The strategy was to use every reasonable excuse to force first-order kinetics on the selected landfill evaluative environment by lumping parameters in 'k'. The primary assumption was that the concentration of second or third reactant is effectively constant and will not change appreciably as the reaction proceeds or reactants other than contaminants are microbial population, sunlight etc. So, it can often circumvent these complex reaction rate equations by expressing them in terms of a pseudo first-order rate reaction. Biodegradation reaction was considered because of the biological conversion of chemical in the landfill. Hydrolysis reaction was considered because of chemical species subjections to rain water and in-situ moisture (leachate). As Rajbandh landfill is an open dumping landfill, there is a considerable exposure to sunlight and atmospheric oxygen. That's why photolysis and oxidation reaction were considered as there is a chance of contaminants to react due to exposure to sunlight and atmospheric and aqueous phase oxygen. Though all these

reactions are simultaneously not applicable for all the contaminants, the combined values of first order reaction rate constant were obtained from Equation 2.

$$K = K_O + K_P + K_H + K_B \quad (2)$$

Where, K is the Combined reaction rate constant (h^{-1}) and K_O , K_P , K_H , K_B are the reaction rate constant for oxidation, photolysis, hydrolysis and biodegradation reactions respectively (h^{-1}).

Although chemical potential has a logarithmic relationship with concentration, Fugacity is linearly related to chemical concentration (C) with a proportionality constant known as Fugacity capacity (Z). It is a function of temperature and natural chemical property. The fugacity capacities (Z) of contaminant at different compartment of the evaluative environment are obtained using the formulas presented in Table 1. The contaminant can be removed from different compartment of the evaluative environment by advection and reaction. Advection is the directed movements of chemical by virtue of its presence in a medium that happens to be flowing. Reaction is the process that alter the chemical nature of the solute and it will exhibit intermedia transport (diffusive flux and material flux process) characteristics between different landfill compartments. To acknowledge advection, reaction and intermedia transport, a term called D value arises which is further used for calculating the rate of intermedia transport, rate of advection and rate of reaction separately.

Table 1: Definition of fugacity capacities

Compartment	Fugacity capacity, Z(mol/m ³ Pa)	Source of parameters
LFG/air (1)	$1/RT^a$	R = 8.314 Pa ³ /mol K T= Absolute temperature (298 K)
Leachate (2)	$1/H$ or C_S/P_S^b	H = Henry's Law Constant (Pa ³ m/mol) C^S = Aqueous Solubility (mol/m ³) P^s = Vapor Pressure (Pa)
Waste (3)	$K_p\rho/H^c$	K_p = Partition Coefficient (L/kg) P = density (kg/L) $K_p = 0.411 \times K_{ow}$ Where, x = Fraction of organic carbon K_{ow} = Octanol-water partition coefficient

^{a, b, c}(Mackay *et al.*, 1985)

D values for intermedia transport are obtained using the formulas presented in Table 2.

Table 2: D values for intermedia transport

From/ To	To/From	D value (mol/Pa. h) ^d	Source of parameters
LFG (1)	Leachate (2)	$D_{12} = \frac{1}{\frac{1}{K_{12}A_{12}Z_1} + \frac{1}{K_{21}A_{12}Z_2}}$	K_{12} = LFG side MTC over leachate (m/h) K_{21} = Leachate side MTC over LFG (m/h) $A_{12} = A_{21}$ = LFG-leachate contact surface area (m ²) Z_1 = Fugacity Capacity at LFG media ((mol/m ³ Pa) Z_2 = Fugacity Capacity at leachate media (mol/m ³ Pa)
LFG (1)	Waste (3)	$D_{13} = \frac{1}{\frac{1}{K_{13}A_{13}Z_1} + \frac{Y_3}{B_3A_{13}Z_3}}$	K_{13} = LFG side MTC over waste (m/h) B_3 = Molecular diffusivity in LFG (m ² /h) $A_{13} = A_{31}$ = LFG-waste contact surface area (m ²) Z_1 = Fugacity Capacity at LFG media (mol/m ³ Pa) Z_3 = Fugacity Capacity at waste media (mol/m ³ Pa) Y_3 = Diffusion path length in waste (m)
Waste (3)	Leachate (2)	$D_{32} = G_w Z_2 + G_s Z_3$	G_w = Leachate runoff rate from waste (m ³ /h) G_s = Solid runoff rate from waste (m ³ /h) Z_2 = Fugacity Capacity at leachate media (mol/m ³ Pa) Z_3 = Fugacity Capacity at waste media (mol/m ³ Pa)

^d(Mackay *et al.*, 1985)

* $D_{ij} = D_{ji}$ as only diffusive transport is considered.

D values for advection can be found using Equation 3.

$$D_A = GZ \quad (3)$$

Where, D_A denotes D values for advection (mol/Pa h), G is advection inflow rate of the contaminant (m³/h) and Z is fugacity capacity (mol/m³ Pa).

D values for reaction can be found using Equation 4.

$$D_R = VKZ \quad (4)$$

Where, D_R denotes D values for reaction (mol/Pa h), V is volume of the compartment (m^3), K is combined reaction rate constant (h^{-1}) and Z is fugacity capacity (mol/ m^3 Pa).

Mass balance equation for LFG, leachate and waste compartment yields the following sets of equations.

$$E_1 + f_2 D_{21} + f_3 D_{31} = f_1 (D_{A1} + D_{R1} + D_{12} + D_{13}) \quad (5)$$

$$E_2 + f_1 D_{12} + f_3 D_{32} = f_2 (D_{A2} + D_{R2} + D_{21}) \quad (6)$$

$$E_3 + f_1 D_{13} = f_3 (D_{A3} + D_{R3} + D_{31} + D_{32}) \quad (7)$$

Where, E_1 is emission rate of contaminant at LFG compartment (mol/h), E_2 is emission rate of contaminant at leachate compartment (mol/h), E_3 is emission rate of contaminant at waste compartment (mol/h).

D_{A1} is D value for advection in LFG (mol/Pa h), D_{A2} is D value for advection in leachate (mol/Pa h), D_{A3} is D value for advection in waste (mol/Pa h), D_{R1} is D values for reaction in LFG (mol/Pa h), D_{R2} is D values for reaction in leachate (mol/Pa h), D_{R3} is D values for reaction in leachate (mol/Pa h), f_1 is fugacity of the contaminant at LFG (Pa), f_2 is fugacity of the contaminant at leachate (Pa), f_3 is fugacity of the contaminant at waste (Pa), D_{12} is the D value for intermedia transport from LFG to leachate, D_{21} is the D value for intermedia transport from leachate to LFG, D_{13} is the D value for intermedia transport from LFG to waste, D_{31} is the D value for intermedia transport from waste to LFG, D_{32} is the D value for intermedia transport from waste to leachate.

Lumping Equation 5, 6 and 7 for the three compartments, a linear matrix can be formed. Solving this matrix, fugacity (Pa) of the contaminant in each of the compartment can be obtained. As concentration is linearly proportional to the fugacity, the concentration of contaminant at different compartment can be found out by Equation 8.

$$C = fZ \quad (8)$$

Where, C denotes Concentration (mol/ m^3), f is the fugacity of contaminant at corresponding compartment (Pa), Z denotes the fugacity capacity of contaminant at that compartment.

The removal rate of contaminants, which are the rate of advection and reaction can be found out by Equation 9 and 10.

$$E_A = D_A f \quad (9)$$

$$E_R = D_R f \quad (10)$$

Where, E_A denotes rate of advection (mol/h), E_R denotes rate of reaction (mol/h), D_A denotes D values for advection (mol/Pa h), D_R denotes D values for reaction (mol/Pa h) and f is the fugacity of contaminant at corresponding compartment (Pa).

Rate of intermedia transport between two compartments can be found out by Equation 11.

$$E_{ij} = D_{ij} f_i - D_{ji} f_j \quad (11)$$

Where, E_{ij} is the rate of intermedia transport from compartment i to compartment j , D_{ij} is the D value for intermedia transport from i to j , D_{ji} is the D value for intermedia transport from j to i and, f_i is the fugacity of contaminant at i compartment (Pa) and f_j is the fugacity of contaminant at j compartment (Pa).

3. MATERIALS AND METHODS

The materials and methods adopted in this study are described in the following articles.

3.1 Description of Study Area

The Khulna city is in the southwestern part of Bangladesh, acknowledged as third highest among ten metropolitan cities of Bangladesh. Geographically, Khulna lies between $22^{\circ}47'16''$ to $22^{\circ}52'0''$ north latitude and $89^{\circ}31'36''$ to $89^{\circ}34'35''$ east longitude (Rafizul and Fahmida, 2019). This city is situated on the bank of Rupsha and Bhairab River. At present, the city is covering an area of 45.65 km^2 containing a population about 1.5 million. The MSW produced by the people of Khulna city is dumped in an open disposal site known as old Rajbandh in order to accumulate and dispose it in the waste disposal site (Pangkaj, 2018). The waste disposal site, Rajbandh is located at the north side of Khulna-Satkhira highway and approximately 8 km far from the city center. Figure 1 depicts the location map of Rajbandh, Khulna, Bangladesh. Khulna city generates approximately 450 tons of MSW every day which increases to approximately 500 tons due to demand and seasonal variation of products (Hasib and Rafizul, 2020). Of the total amount, only 250-270 tons of MSW are

dumped into the open dumping ground at Rajbandh, Khulna (Khan *et al.*, 2015). Therefore, open dumping site at old Rajbandh, Khulna has been selected as a case study.

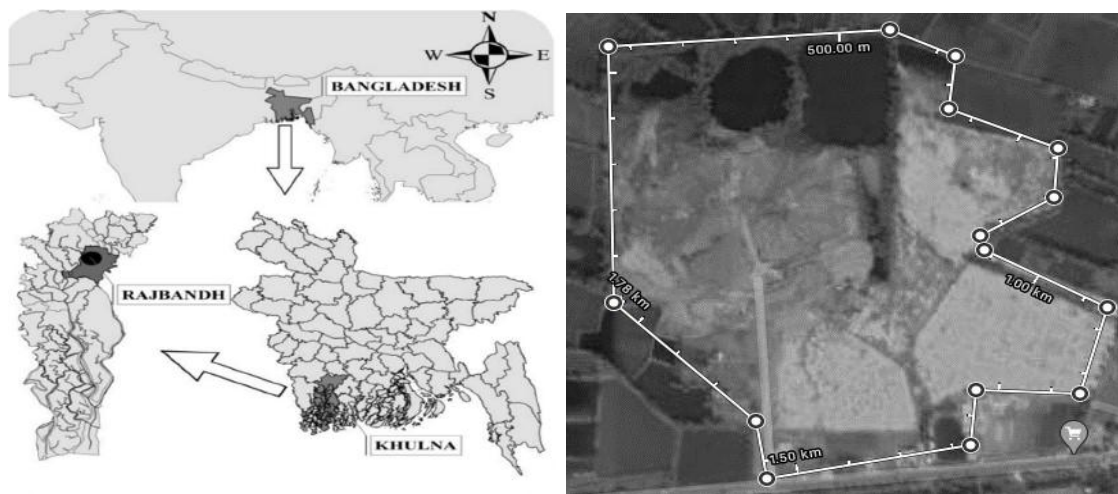


Figure 1: Map showing selected open dumping site at old Rajbandh, Khulna, Bangladesh

3.2 Evaluative Landfill Environment

The dumping site of old Rajbandh, Khulna was taken as the evaluative environment which allows for three phases: landfill gas (LFG), water (leachate) and solid waste for processes of gas and water flux through the system being represented. The entire dumping site was considered as a single cell of MSW. This choice was influenced by the availability of data on the site-specific properties. The dimensions of the selected area of landfill (Figure 1) and characteristics of the evaluative environment are reported in Table 3. The volumetric composition of LFG/air, leachate and waste of the evaluative environment were extracted using data regarding leachate content, LFG and leachate flow rate and total volume of the corresponding landfill (Islam, 2014) and were found 10%, 65% and 25% of the total cell volume, respectively. This implies the volume of $2.17\text{E}+04\text{ m}^3$, $1.27\text{E}+05\text{ m}^3$ and $6.83\text{E}+04\text{ m}^3$ for LFG, leachate and waste compartment respectively.

Table 3: Dimension of evaluative environment

Parameter	Value
Cell area	$5.42\text{E}+04\text{ m}^2$
Cell depth	4 m^e
Cell volume	$2.17\text{E}+05\text{ m}^3$
Waste density	$1.00\text{E}+03\text{ kg/m}^3.f$
Waste deposition rate	260 ton/day^j

^f(Islam, 2014); ^j(Khan *et al.*, 2015)

3.3 Chemical Input

Environmental fate of contaminants is intimately connected with their physiochemical properties as well as environmental properties in the specific study areas, so model input parameters need to be updated when analysing different contaminants in different study areas (Mackay *et al.*, 1985). There are variabilities and uncertainties associated with chemical input parameters due to lack of local environmental data. For this reason, typical values from (Mackay *et al.*, 1985) were used as the chemical input parameters such as emission rates, reaction rate constants, intermedia transport parameters depicted in Tables 4, 5 and 6 respectively.

The emission rate for all contaminants in the evaluative environment was considered 1 mol/h and it distributes in different compartments of the evaluative environment according to their nature. Due to lack of local environmental data, no background concentration of the contaminants was considered. In Monte Carlo simulation using @RISK 7.6, these chemical input parameters were varied in nature of a normal distribution curve (except emission rates) with suitable mean and standard deviation values supporting the literatures. The emission rates were varied as uniform distribution for ensuring the constant continuous emission in the corresponding compartment of evaluative environment.

Table 4: Chemical properties and emission rates

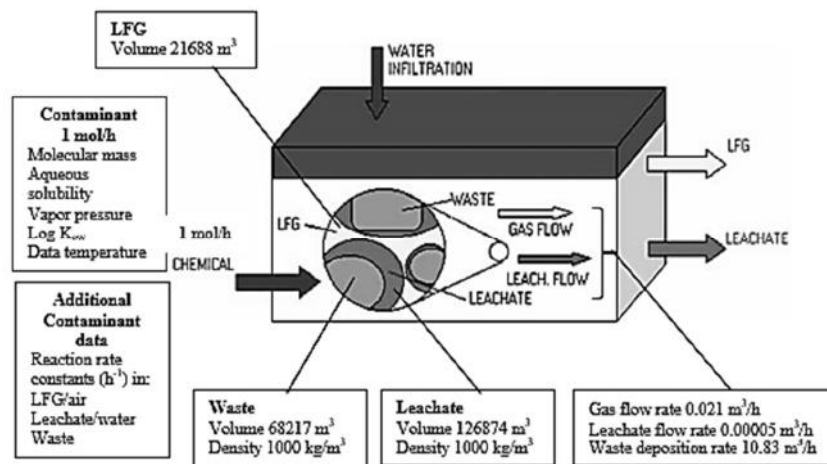
Chemical Name	MW (g/mol)	C ^s (g/m ³)	P ^s (Pa)	Log K _{ow}	Emission Rate (mol/h)		
					LFG/air	Leachate	Waste/soil
Trichloroethylene	1.3E+02	1.8E+03	1.1E+03	2.3E+00	9.0E-01	5.0E-02	5.0E-02
Atrazine	2.2E+02	3.3E+01	3.0E-06	2.3E+00	0.0E+00	1.0E-01	9.0E-01
m-Xylene	1.1E+02	1.6E+02	1.1E+03	3.2E+00	9.3E-01	4.3E-02	2.7E-02
Benzene	7.8E+01	1.8E+03	1.3E+04	2.1E+00	0.0E+00	1.0E+00	0.0E+00

Table 5: First order reaction rate constants

Chemicals	Reaction Rate Constant, K (h ⁻¹)						
	Photolysis	Oxidation		Hydrolysis		Biodegradation	
	LFG/air	LFG/air	Leachate	Waste/soil	Leachate	Leachate	Waste/soil
Trichloroethylene	0.0E+00	7.2E-03	0.0E+00	0.0E+00	9.0E-05	0.0E+00	0.0E+00
Atrazine	3.7E-02	0.0E+00	0.0E+00	1.4E-04	0.0E+00	3.9E-03	8.0E-05
m-Xylene	0.0E+00	0.0E+00	1.4E-07	0.0E+00	0.0E+00	1.0E-03	0.0E+00
Benzene	8.6E-04	1.8E-04	0.0E+00	0.0E+00	0.0E+00	4.6E-03	0.0E+00

Table 6: Intermedia transport parameters

Parameter	Symbol	Value	Justification/Reference
LFG side MTC over leachate	K ₁₂	10 m/h	Taken from (Mackay <i>et al.</i> , 1985)
Leachate side MTC over LFG	K ₂₁	0.1 m/h	
LFG side MTC over waste	K ₁₃	10 m/h	
Molecular diffusivity in LFG	B ₃	0.04 m ² /h	
LFG-Leachate contact area	A ₁₂	54200 m ²	Adapted to evaluative environment
LFG-waste contact area	A ₁₃	54200 m ²	
Leachate runoff rate from waste	G _w	5.00E-05 m ³ /h	
Solid runoff rate from waste	G _S	1.08E+01 m ³ /h	
Diffusion path length in waste	Y ₃	2 m	

**Figure 2:** Evaluative environment for Level III Fugacity model (Source: Shafi *et al.*, 2006)

3.4 Fugacity Modelling

Level III fugacity calculations illustrate the partitioning behaviour of contaminants in the evaluative environment of landfill. Level III model accounts for steady state but non-equilibrium flow system i.e. the amount of contaminant entering the environment is mass balanced by the amount lost to flow, reaction or degradation and intermedia transport (Shafi *et al.*, 2006). The entire phenomenon is briefly illustrated in Figure 2 (Shafi *et al.*, 2006) where contaminant is introduced in the evaluative landfill environment. It partitions among the LFG, leachate and waste compartment and after certain residence time, the contaminant is removed by flow and reaction. The characteristics of the system are presented in Table 7. Using Equations (1) to (11), the

behaviour such as concentration, fugacity, mass distribution, rate of advection, rate of reaction, rate of intermedia transport, residence time etc. of specific contaminants in LFG, leachate and waste compartment were characterised.

Table 7: Parameters for Level III Fugacity model calculation

Parameter	Value	Justification/Reference
Landfill gas flow rate	2.10E-02 m ³ /h	Adapted to evaluative environment ^k
Leachate flow rate	5.00E-05 m ³ /h	Adapted to evaluative environment ^l

^{k, l} (Islam, 2009, 2014)

3.5 Monte Carlo Simulation

As models are only approximation of the contaminants actual behaviour in real environment, communicating the uncertainties associated with the model is crucial (Kilic and Aral, 2008). Monte Carlo simulation (MCS) is a great tool for quantifying uncertainties in fugacity model. A typical MCS calculates the model hundreds of times and each time it uses a randomly selected values for the input parameters. When the simulation is complete, it has large number of results from the model, each based on random input values. These results describe the likelihood, or probability of reaching various results in the model (Pangkaj, 2018). It also justifies the outcomes of the model. In this study, chemical input parameters such as aqueous solubility, vapor pressure, first order reaction rate constants, octanol water partition coefficients etc. of contaminants in different compartments have been assigned lognormal distribution as it has a positive state space. The emission rates were assigned uniform distribution with support of literature. They were assigned with suitable mean and standard deviation values supporting the literatures. For each contaminant, the highest concentration in respective compartment was chosen as the output parameter of the simulation. A total of 10000 trials were performed using @RISK 7.6 for the MCS runs with 10000 random variables created for the overall aqueous solubility, vapor pressure, first order reaction rate constants, octanol water partition coefficients and contaminant emission rates. The simulation outcomes were used to check the level III Fugacity model outcomes and the most sensitive parameters were recognized from tornado charts.

4. RESULTS AND DISCUSSION

The findings from level III Fugacity model and Monte Carlo simulation are presented and hence discussed in the following articles.

4.1 Level III Fugacity Modelling

The level III Fugacity model was implemented to characterize the selected contaminants emitted from MSW landfill and the findings are highlighted in the following sections.

4.1.1 Trichloroethylene

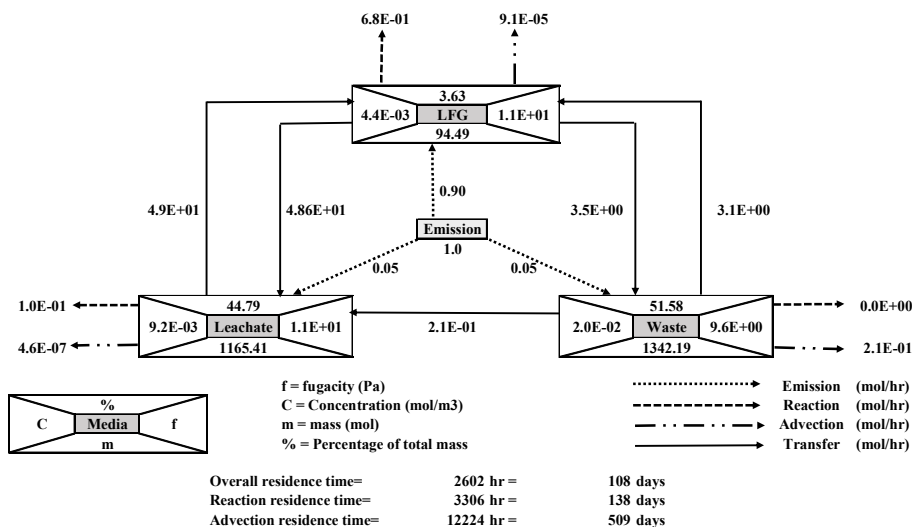


Figure 3: Level III distribution of Trichloroethylene in landfill evaluative environment

Figure 3 shows the behaviour of Trichloroethylene in landfill evaluative environment according to Level III Fugacity model. Five intermedia transport ways were considered which are LFG to leachate, leachate to LFG, waste to LFG, LFG to waste and waste to leachate (Figure 3). Intermedia transport rate of trichloroethylene from LFG to leachate was found $4.86E+01$ mol/hr while leachate to LFG was found $4.9E+01$ mol/hr. So, net intermedia transport rate between LFG and leachate was $1.6E-01$ mol/hr from leachate to LFG compartment (Figure 3). Transport rate of trichloroethylene from LFG to waste was found $3.5E+00$ mol/hr while waste to LFG was found $3.1E+00$ mol/hr. So, net intermedia transport rate between LFG and waste was $3.8E-01$ mol/hr from LFG to waste compartment. Intermedia transport rate from waste to leachate was found $2.1E-01$ mol/hr (Figure 3). Advection residence time was 509 days and reaction residence time were 138 days. But the overall persistence time of Trichloroethylene was 108 days (Figure 3).

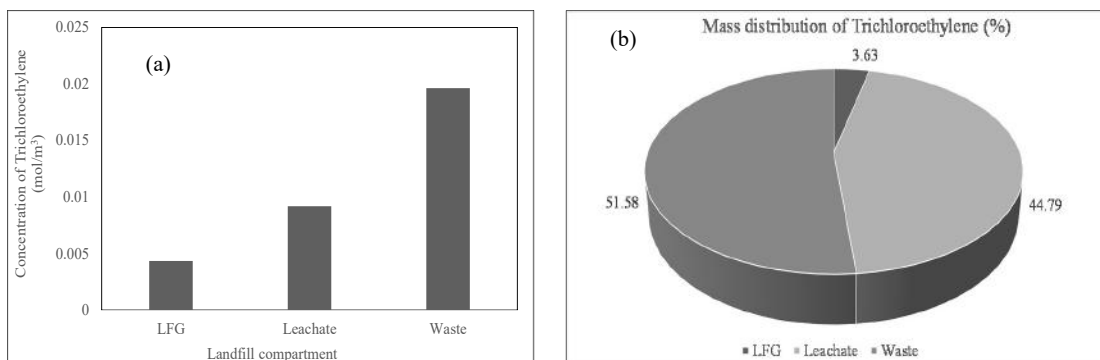


Figure 4: Behaviour of Trichloroethylene in evaluative environment (a) concentration and (b) mass distribution

The behaviour of Trichloroethylene interms of concentrations and mass in landfill compartment is shown in Figure 4. The modelled concentration of Trichloroethylene was found $4.4E-03$, $9.2E-03$ and $2.0E-02$ mol/m³ in LFG, leachate and waste compartment, respectively (Figure 4a). Highest concentration was found in the waste compartment (Figure 4a). Most of the mass (51.58%) was found in waste compartment (Figure 4b). About 45% of the total mass was found in leachate compartment and the rest was in LFG compartment.

The behaviour depicted in Figure 5 showed that about 68.23% of the total emitted Trichloroethylene was removed by oxidation reaction in LFG, 21.28% by advection in waste and the remaining 10.49% was removed by hydrolysis reaction in landfill leachate. About 79% of total emitted amount was removed by reaction. So, advection was relatively unimportant compared to reaction as a removal mechanism for Trichloroethylene.

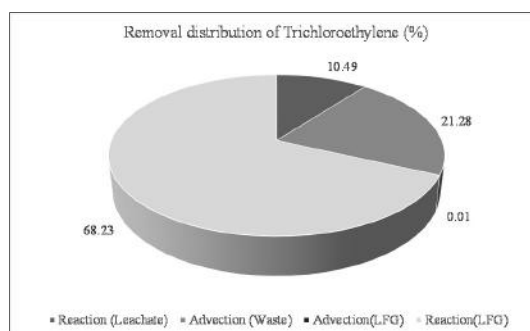


Figure 5: Removal distribution of Trichloroethylene in evaluative environment

4.1.2 Atrazine

The distribution of Atrazine in the evaluative environment is shown in Figure 6. Intermedia transport rate of atrazine from LFG to leachate was found $2.16E-05$ mol/hr, while, leachate to LFG was found $3.22E-06$ mol/hr. So, net intermedia transport rate between LFG and leachate was $1.84E-05$ mol/hr from LFG to leachate compartment (Figure 6). Transport rate of atrazine from LFG to waste was found $2.16E-05$ mol/hr while waste to LFG was found $4.01E-05$ mol/hr. So, net intermedia transport rate between LFG and waste was $1.84E-05$ mol/hr from waste to LFG compartment which is a very little amount (Figure 6). Intermedia transport rate from waste to leachate was found $2.67E-01$ mol/hr (Figure 6). Advection residence time was 278 days and reaction residence time were 101 days. But the overall persistence time of Atrazine was 74 days (Figure 6).

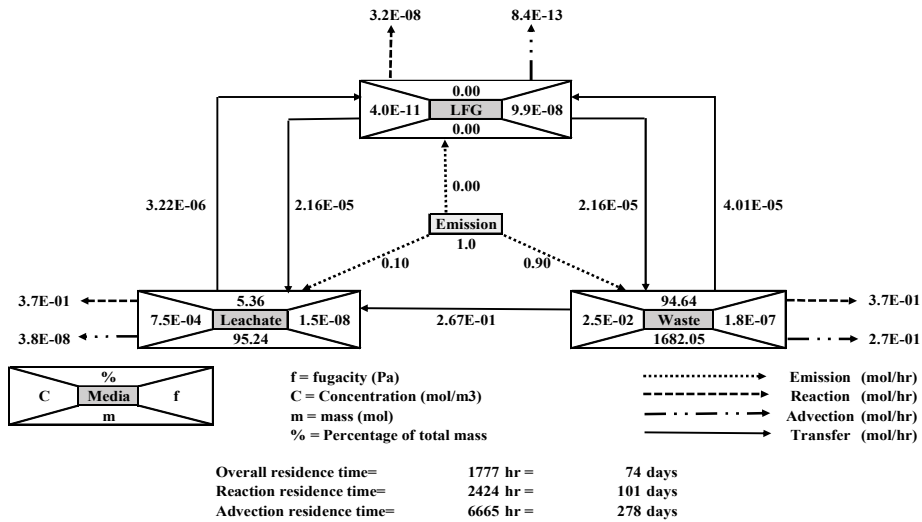


Figure 6: Level III distribution of Atrazine in landfill evaluative environment

The behaviour of Atrazine interms of concentrations and mass in landfill compartment is shown in Figure 7. The concentration of Atrazine was found approximately 4.0E-11, 7.5E-04 and 2.5E-02 mol/m³ in LFG, leachate and waste compartment, respectively (Figure 7a). Negligible amount of concentrations was found in LFG compartment due to no emission of it in LFG and very little intermedia intermedia transfer rate in that compartment. About 94.64% of total mass was found in waste and remaining 5.36% was in leachate (Figure 7b).

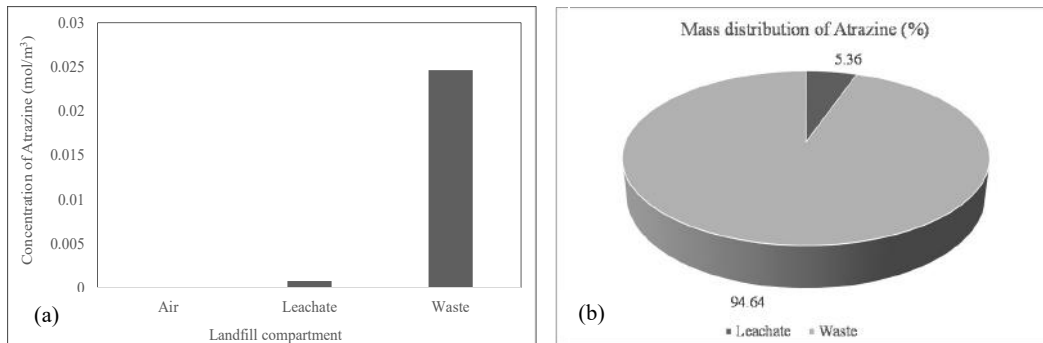


Figure 7: Behaviour of Atrazine in evaluative environment (a) concentration and (b) mass distribution

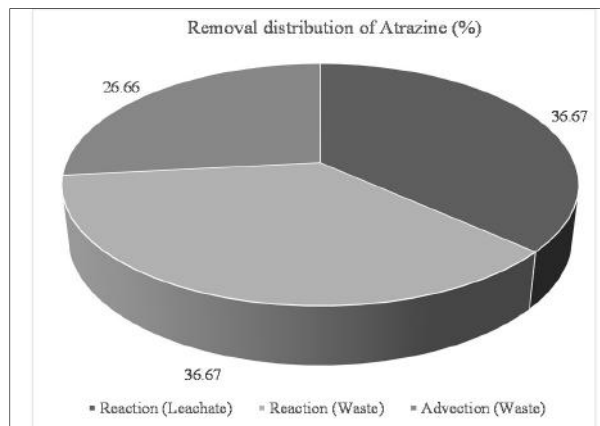


Figure 8: Removal distribution of Trichloroethylene in evaluative environment

The illustration in Figure 8 showed that about 36.67% Atrazine was removed by reaction (photolysis and hydrolysis) in waste, 26.66% by advection in waste and the remaining 36.67% of atrazine was removed by

reaction (biodegradation) in leachate. So, reaction (photolysis, hydrolysis, and biodegradation) was the main removal mechanism for Atrazine as about 73% of the total emitted amount was removed by reaction.

4.1.3 m-Xylene

Figure 9 shows the behaviour of m-Xylene in landfill evaluative environment according to Level III Fugacity model. Intermedia transport rate of m-Xylene from LFG to leachate was found $2.26E+01$ mol/hr while leachate to LFG was found $2.25E+01$ mol/hr. So, net intermedia transport rate between LFG and leachate was $7.88E-02$ mol/hr from LFG to leachate compartment (Figure 9). Transport rate of m-Xylene from LFG to waste was found $1.64E+00$ mol/hr while waste to LFG was found $7.91E-01$ mol/hr. So, net intermedia transport rate between LFG and waste was $8.51E-01$ mol/hr from LFG to waste compartment (Figure 9). Intermedia transport rate from waste to leachate was found $4.39E-01$ mol/hr (Figure 9). Advection residence time was 317 days and reaction residence time was 248 days. But the overall persistence time of M-Xylene was 139 days (Figure 9).

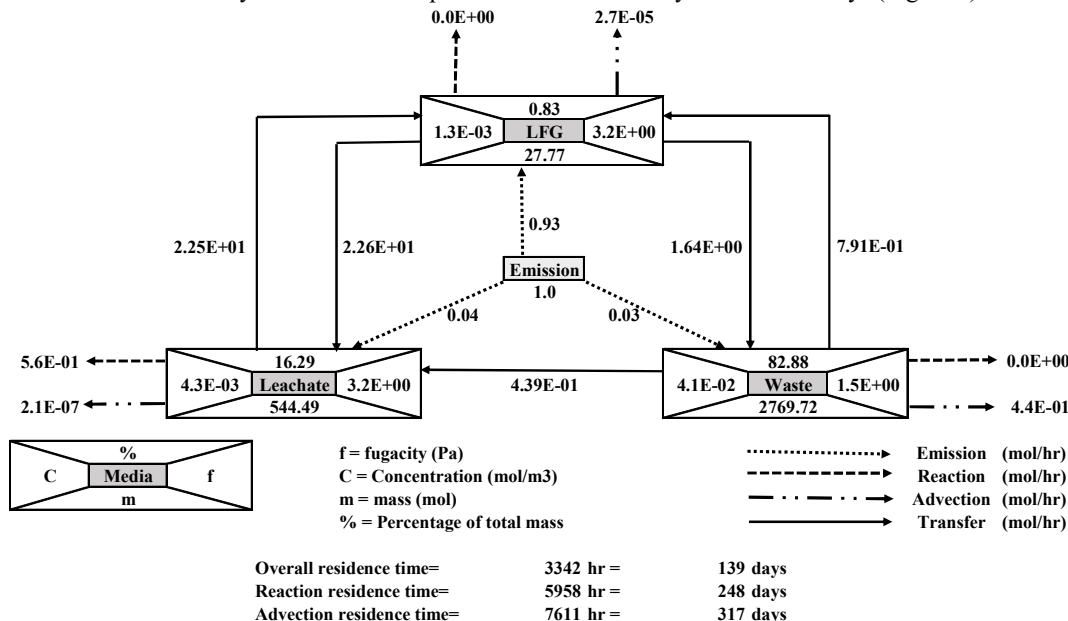


Figure 9: Level III distribution of m-Xylene in landfill evaluative environment

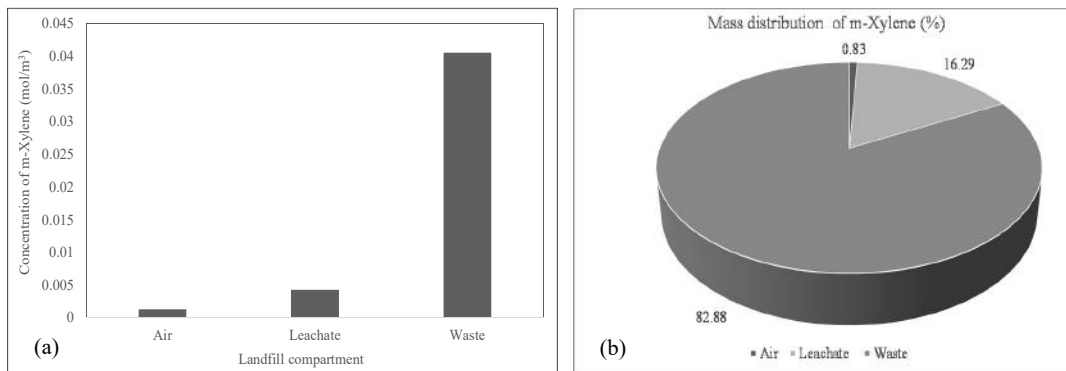


Figure 10: Behaviour of m-Xylene in evaluative environment (a) concentration and (b) mass distribution

The behaviour of m-Xylene interms of concentrations and mass in landfill compartment is shown in Figure 10. The concentration of m-Xylene was found $1.3E-03$ mol/m³, $4.3E-03$ mol/m³ and $4.1E-02$ mol/m³ in LFG, leachate and waste compartment, respectively (Figure 10a). High concentration (Figure 10a) was found in the waste compartment. Most of the mass (82.88%) was found in waste compartment (Figure 10b). About 16.29% of the total mass was found in leachate compartment and the rest was in LFG compartment (Figure 10b). Figure 11 showed that about 56% of the total emitted m-Xylene was removed by biodegradation and oxidation reaction in leachate and the remaining 44% by advection in waste. So, reaction was the main removal mechanism for m-Xylene.

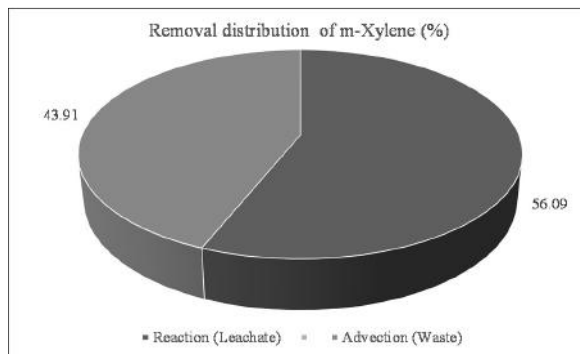


Figure 11: Removal distribution of m-Xylene in evaluative environment

4.1.4 Benzene

Figure 12 shows the behaviour of Benzene in landfill evaluative environment according to Level III Fugacity model. Intermedia transport rate of benzene from LFG to leachate was found $8.25E+00$ mol/hr while leachate to LFG was found $8.31E+00$ mol/hr. So, net intermedia transport rate between LFG and leachate was $5.90E-02$ mol/hr from leachate to LFG compartment (Figure 12). Transport rate of benzene from LFG to waste was found $6.1E-01$ mol/hr while waste to LFG was found $5.5E-01$ mol/hr. So, net intermedia transport rate between LFG and waste was $5.2E-02$ mol/hr from LFG to waste compartment. Intermedia transport rate from waste to leachate was found $2.6E-02$ mol/hr (Figure 12). Advection residence time was 599 days and reaction residence time were 16 days. Overall persistence time of Benzene was 16 days (Figure 12).

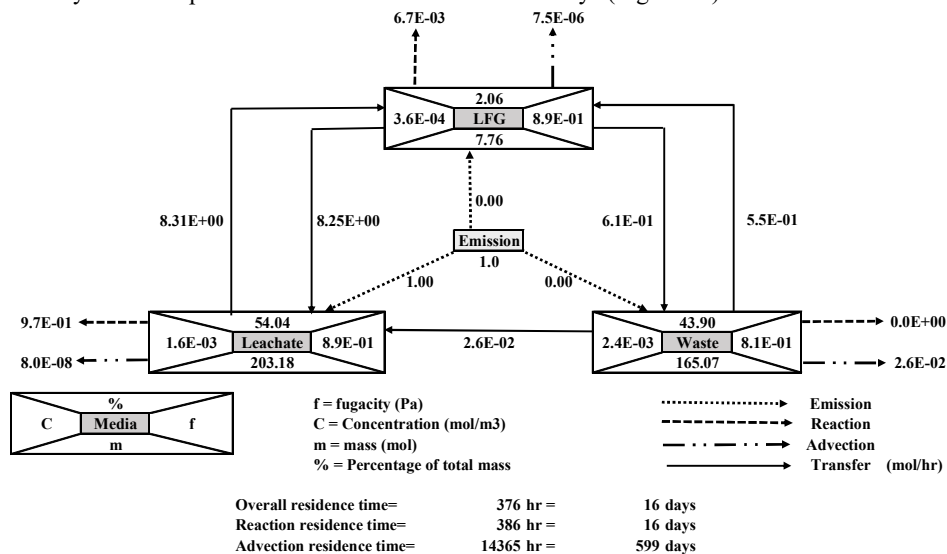


Figure 12: Level III distribution of Benzene in landfill evaluative environment

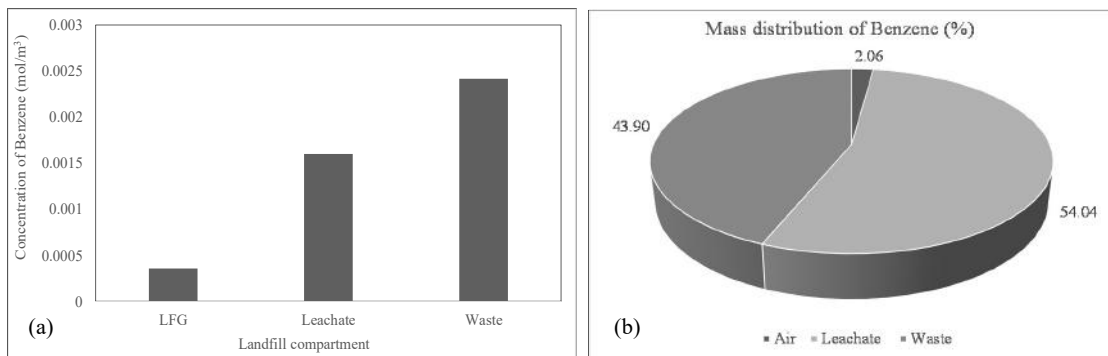


Figure 13: Behaviour of Benzene in evaluative environment (a) concentration and (b) mass distribution

The behaviour of Benzene in terms of concentrations and mass in landfill compartment is shown in Figure 13. The modelled concentration of Benzene was found $3.6\text{E-}04$, $1.6\text{E-}03$ and $2.4\text{E-}03$ mol/m³ in LFG, leachate and waste compartment, respectively (Figure 13a). Highest concentration was found in the waste compartment (Figure 13a). Most of the mass (54%) was found in leachate compartment (Figure 13b) because of its high volume. About 44% of the total mass was found in waste compartment and the rest 2% was in LFG compartment (Figure 13b). The behaviour depicted in Figure 14 showed that about 96.71% of the total emitted Benzene was removed by photolysis and biodegradation reaction in leachate, 2.68% by advection in waste and the remaining 0.67% was removed by photolysis reaction in LFG. About 98% of total emitted amount was removed by reaction. So, advection was relatively unimportant compared to reaction as a removal mechanism for Benzene.

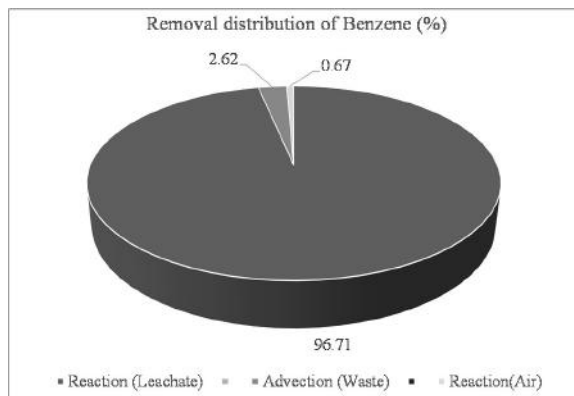


Figure 14: Removal distribution of Benzene in evaluative environment

4.2 Monte Carlo Simulation

To fit a uniform distribution of the emission rates and normal distribution of other chemical input parameters, this highest concentration was compiled using @RISK 7.6. The MCS was run for 10000 trials with 10000 random variables for the input parameters of Fugacity model. The most sensitive parameter in MCS runs was found by varying each of the input parameter within the assigned normal or uniform distribution curve and keeping the other parameters at their static values.

4.2.1 Trichloroethylene

Highest concentration of Trichloroethylene was found in waste compartment with a value of $2.0\text{E-}02$ mol/m³. In Figure 15, the height of the bars (y axis) represents the relative frequency of this concentration and the spread of the bars represents (x axis) the varying amount of this concentration. From Figure 15, it is seen that the concentration of Trichloroethylene in waste media ranges from $2.8\text{E-}03$ to $9.6\text{E-}03$ mol/m³ and $3.4\text{E-}02$ to $5.6\text{E-}02$ mol/m³ for the 5th and 95th percentile, respectively. The concentration of $9.6\text{E-}03$ mol/m³ responds to 5th percentile and $3.4\text{E-}02$ mol/m³ responds to 95th percentile. So approximately, 90% of the concentration is likely to be exist between $9.6\text{E-}03$ to $3.4\text{E-}02$ mol/m³ with a mean value of $2.04\text{E-}03$ mol/m³. The modelled concentration was also found within this range.

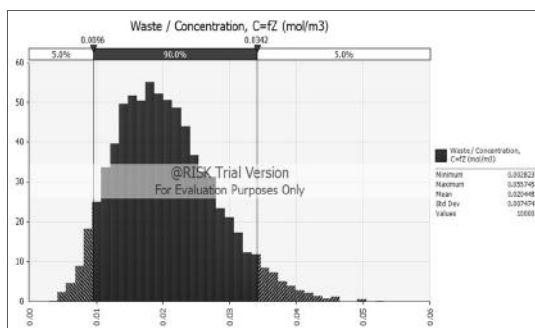


Figure 15: Monte Carlo simulation run for Trichloroethylene

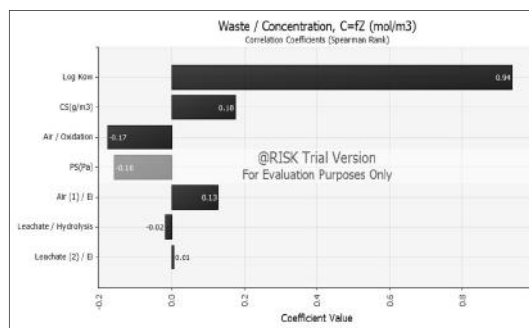


Figure 16: Sensitivity analysis for concentration of Trichloroethylene in waste compartment

Sensitivity analysis was performed to determine which chemical input variables had the greatest impact on the concentration of Trichloroethylene in waste compartment. The results of sensitivity analysis are shown in Figure

16 in the form of tornado plot illustrating the Spearman rank order correlation coefficients. It was also found that octanol-water partition coefficient was the most sensitive parameter for variation of this concentration in Monte Carlo runs (Figure 16). It has a higher positive correlation coefficients which indicates that the concentration will increase if octanol-water partition coefficient increases and vice versa. Figure 16 also indicates that octanol-water partition coefficient, aqueous solubility, oxidation reaction rate constant in LFG, vapor pressure and emission rate of Trichloroethylene in LFG shouldn't be treated as a single valued input variable. The concentration increases if the oxidation reaction rate constant in LFG compartment and vapor pressure decreases and vice versa as it have a negative correlation coefficient (Figure 16). Concentration also increases if the emission rate of Trichloroethylene in LFG media increases and vice versa (Figure 16).

4.2.2 Atrazine

Atrazine had highest concentration in waste compartment with a value of $2.5E-02 \text{ mol/m}^3$. In Figure 17, the height of the bars (y axis) represents the relative frequency of this concentration and the spread of the bars represents (x axis) the varying amount of this concentration. From Figure 17, it is seen that the concentration of Atrazine in waste media ranges from $2.00E-02$ to $2.21E-02 \text{ mol/m}^3$ and $2.72E-02$ to $2.95E-02 \text{ mol/m}^3$ for the 5th and 95th percentile, respectively. The concentration of $2.21E-02 \text{ mol/m}^3$ responds to 5th percentile and $2.72E-02 \text{ mol/m}^3$ responds to 95th percentile. So, approximately 90% of the concentration is likely to be exist between $2.21E-02$ to $2.72E-02 \text{ mol/m}^3$ with a mean value of $2.46E-02 \text{ mol/m}^3$. The modelled concentration was also found within this range.

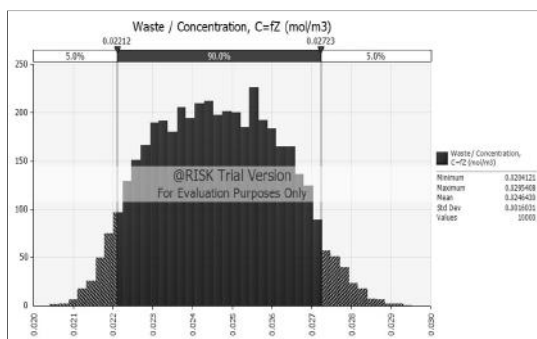


Figure 17: Monte Carlo simulation run for Atrazine

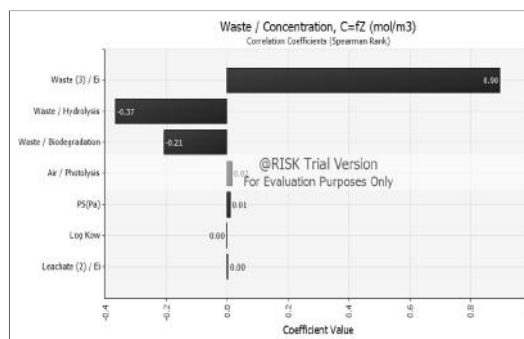


Figure 18: Sensitivity analysis for concentration of Atrazine in waste compartment

The results of sensitivity analysis are shown in Figure 18 in the form of tornado plot illustrating the Spearman rank order correlation coefficients. It was also found that emission rate of atrazine in waste compartment was the most sensitive parameter for variation of this concentration in Monte Carlo runs (Figure 18). It has a higher positive correlation coefficients which indicates that the concentration will increase if emission rate at that compartment increases and vice versa. Figure 18 also indicates that it is reasonable to consider octanol-water partition coefficient, aqueous solubility, photolysis reaction rate constant in LFG, vapor pressure and emission rate of atrazine in leachate as a single valued input variable. Figure 18 also shows that the concentration decreases if the hydrolysis and biodegradation reaction rate constant in waste compartment increases and vice versa as it have a negative correlation coefficient.

4.2.3 m-Xylene

m-Xylene had highest concentration in LFG compartment with a value of $4.1E-02 \text{ mol/m}^3$. In Figure 19, the height of the bars (y axis) represents the relative frequency of this concentration and the spread of the bars represents (x axis) the varying amount of this concentration. From Figure 19, it is seen that the concentration of m-Xylene in LFG media ranges from $6.0E-03$ to $2.4E-02 \text{ mol/m}^3$ and $5.1E-02$ to $6.4E-02 \text{ mol/m}^3$ for the 5th and 95th percentile respectively. $2.4E-02 \text{ mol/m}^3$ responds to 5th percentile and $5.1E-02 \text{ mol/m}^3$ responds to 95th percentile. So, approximately 90% of the concentration is likely to be exist between $2.4E-02$ to $5.1E-02 \text{ mol/m}^3$ with a mean value of $3.91E-02 \text{ mol/m}^3$. The modelled concentration was also found within this range.

The results of sensitivity analysis are shown in Figure 20 in the form of tornado plot illustrating the Spearman rank order correlation coefficients. It was also found that octanol-water partition coefficient of m-Xylene was the most sensitive parameter for variation of this concentration in Monte Carlo runs (Figure 20). It has a higher positive correlation coefficients which indicates that the concentration will increase if emission rate at that compartment increases and vice versa. Figure 20 also indicates that it is reasonable to consider aqueous solubility, oxidation reaction rate constant in leachate, vapor pressure and emission rate of m-Xylene in waste

as a single valued input variable. Figure 20 also shows that the concentration decreases if the biodegradation reaction rate constant in leachate compartment increases and vice versa as it have a negative correlation coefficient. Concentration also increases if the emission rate of m-Xylene in LFG media increases and vice versa (Figure 20).

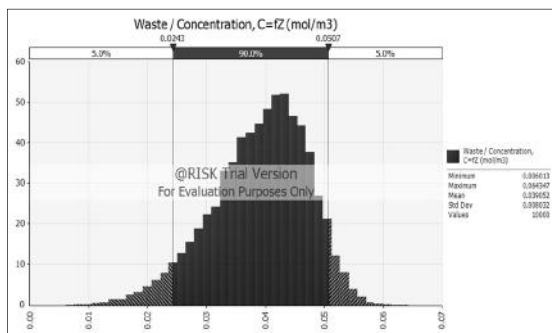


Figure 19: Monte Carlo simulation run for m-Xylene

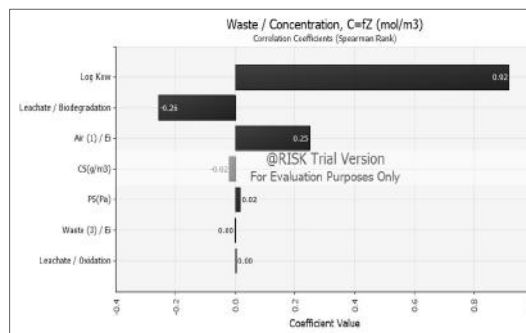


Figure 20: Sensitivity analysis for concentration of m-Xylene in waste compartment

4.2.4 Benzene

Highest concentration of benzene was found in waste compartment with a value of $2.4E-03 \text{ mol/m}^3$. In figure 21, the height of the bars (y axis) represents the relative frequency of this concentration and the spread of the bars represents (x axis) the varying amount of this concentration. From Figure 21, it is seen that the concentration of benzene in waste media ranges from $4.3E-04$ to $1.1E-03 \text{ mol/m}^3$ and $4.6E-03$ to $9.1E-03 \text{ mol/m}^3$ for the 5th and 95th percentile, respectively. The concentration of $1.1E-03 \text{ mol/m}^3$ responds to 5th percentile and $4.6E-03 \text{ mol/m}^3$ responds to 95th percentile. So, approximately 90% of the concentration is likely to be exist between $1.1E-03$ to $4.6E-03 \text{ mol/m}^3$ with a mean value of $2.5E-03 \text{ mol/m}^3$. The modelled concentration was also found within this range.

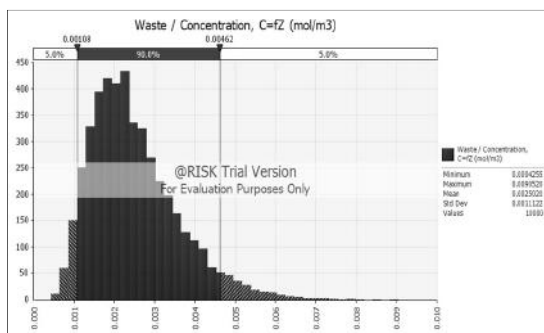


Figure 21: Monte Carlo simulation run for Benzene

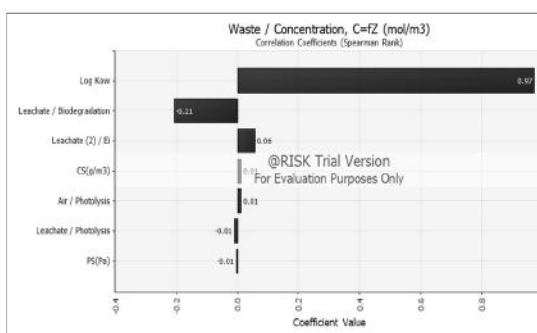


Figure 22: Sensitivity analysis for concentration of Benzene in waste compartment

The results of sensitivity analysis are shown in Figure 22 in the form of tornado plot illustrating the Spearman rank order correlation coefficients. It was also found that octanol-water partition coefficient was the most sensitive parameter for variation of this concentration in Monte Carlo runs (Figure 22). It has a higher positive correlation coefficients which indicates that the concentration will increase if octanol-water partition coefficient increases and vice versa. Figure 22 also indicates that octanol-water partition coefficient, biodegradation reaction rate constant in leachate and emission rate of Benzene in leachate shouldn't be treated as a single valued input variable. The concentration increases if the biodegradation reaction rate constant in leachate compartment decreases and vice versa as it have a negative correlation coefficient (Figure 22). Concentration also increases if the emission rate of Benzene in leachate media increases and vice versa.

5. STUDY LIMITATIONS

Lack of site-specific data regarding input parameters was one of the major limitations of this study. It was important in terms of emission rates, but also for the volumetric composition of the LFG, leachate and waste that are required to create the evaluative environment. Due to lack of local environmental data, representative mean values of input parameters from (Mackay *et al.*, 1985) were considered including emission rate, aqueous solubility, vapor pressure, octanol-water partition coefficient and so on. Though the physical properties

considered in this study were site specific (Table 3 and 7), in literature, many of the researchers used the same data presented in Table 4, 5 and 6 for evaluating the contaminants fate as there is a lack of site-specific chemical input parameters (Linyu Xu *et al.*, 2015). In addition, for simplicity, LFG and leachate was treated as having the same input parameters of air and water, respectively, though they would express their own complex characteristics in the real environment as they generate and migrate through the waste. A significant limitation was choosing the Level III Fugacity approach rather than Level IV approach. Level III Fugacity approach doesn't address the time dependent distribution of the contaminants between the compartments of the evaluative environment. Due to lack of data, Level III approach was chosen and it was not capable of representing the actual complex behaviours that contaminants were undergoing in the landfill environment. Another limitation is evaluating the model at standard temperature of 25°C though landfill will undergo diurnal, seasonal and microbiologically induced temperature changes through various stages of its lifetime (Shafi *et al.*, 2006). Temperature is influential for factors like aqueous solubility, vapor pressure and octanol-water partition coefficient. But Fugacity model cannot address this problem without running multiple simulations.

6. CONCLUSIONS

In modelled environment, all contaminants had the highest concentration in municipal solid waste of Khulna landfill. Reaction was found as the main removal mechanism for all contaminants, while, advection was relatively unimportant as a removal mechanism of organic contaminants under level III Fugacity approach. It was found that m-Xylene was the highly persistent organic contaminant as it spends the highest amount of time in the modelled landfill environment. As standard data set regarding Fugacity model for the selected landfill site was sparse, Monte Carlo simulation justified the model outcomes primarily and reduced the variability and uncertainty associated with input parameters addressed in the model. There is an explicit need for precise site-specific chemical input parameters and emission rates to increase the efficiency of this model. It is essential to know the behaviour of potential harmful contaminants for assessing human health hazards from landfill site. The outcome of level III Fugacity model like mass, concentrations, etc. of organic contaminants generated from a selected landfill which will further be helpful for evaluating health hazards from landfill site at Khulna.

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