ASSESSMENT OF THE BEHAVIOUR OF CONTAMINANTS OF A WASTE DISPOSAL SITE AT KHULNA IN BANGLADESH USING FUGACITY MODEL

MD. Nafees Fuad Rafi*[1], Sk. Atikur Rahman[1] and Islam M. Rafizul[2]

[1]*Under-graduate student, Department of Civil Engineering, Khulna University of Engineering & Technology, Khulna-9203, BANGLADESH, e-mail: rafi1501008@stud.kuet.ac.bd*
[1]*Under-graduate student, Department of Civil Engineering, Khulna University of Engineering & Technology, Khulna-9203, BANGLADESH, e-mail: rahman1501088@stud.kuet.ac.bd*
[2]*Professor, Department of Civil Engineering, Khulna University of Engineering & Technology, Khulna-9203, BANGLADESH, e-maill: imrafizul@yahoo.com*

***Corresponding Author**

ABSTRACT

Landfill wastes contain various kind of toxic contaminants such as Atrazine, Trichloroethylene, m-Xylene etc. This study was undertaken to assess the behaviour of such contaminants through Level II Fugacity model in different components like landfill gas (LFG), leachate and waste itself due to emission from a selected waste disposal site at old Rajbandh, Khulna, Bangladesh. In this study, an evaluative environment was considered using fictitious but realistic properties like volume, temperature and composition of the waste disposal site at Rajbandh for implementing the Fugacity Model. The model finally shows the concentration, mass distribution, rate of reaction, rate of advection etc. of the selected chemical compounds in different waste media. The concentration of Trichloroethylene in landfill gas (Air), leachate and waste media was found 5.70E-03 mol/m^3, 4.40E-03 mol/m^3 and 4.60E-03 mol/m^3 respectively modelled under Fugacity approach. An uncertainty analysis was also conducted using Monte Carlo simulation (MCS) in order to account the variability and uncertainty of the model inputs as well as to observe its effect on the model outputs. The most likely range (90%) of the concentration of Trichloroethylene, Atrazine and m-Xylene was found 4.53E-03 - 7.29E-03 mol/m^3, 2.90E-02 - 4.14E-02 mol/m^3 and 3.70E+01 - 5.16E+01 mol/m^3 in LFG, waste and LFG media, respectively, through MCS using @RISK 7.6 with 5000 iterations. The concentrations from Fugacity model for these contaminants also found within these ranges which justified the model outcomes. The simulation from MCS also revealed the input parameters which had the most impact on behaviour of the contaminants. It was found that, oxidation reaction rate constant at landfill gas, emission rate of Atrazine in waste media and emission rate of m-Xylene in LFG media were the most important parameter for characterizing the concentration of Trichloroethylene, Atrazine and m-Xylene in LFG, waste and LFG media, respectively. The study results instruct the health risk management of landfill and help future health risk predcition and control. This study highlights the need (i) for accurate emission rate data of the contaminants in different compartments of evaluative environment and (ii) accurate site specific properties (density, aqueous solubility, temperature, vapor pressure, first order reaction rate constants and octanol-water partition coefficients) of the contaminants in different compartment corresponding to the real environment.

Keywords: *Fugacity model, Monte Carlo simulation, Contaminants, Advection, Reaction.*

1. INTRODUCTION

1.1 Background

Landfill is a unit operation system for solid waste 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 C. et al.,1999). Rajbandh landfill is the main waste disposal site for the Khulna city. The city generates approximately 450 tons of municipal solid waste (MSW) every day which increases to approximately 500 tons due to demand and seasonal variation of products. Of the total amount, only 250-270 tons of MSW are dumped into the open dumping ground at Rajbandh, Khulna (Khan M.S. et al., 2015). As these wastes decomposes, various types of toxic LFG and toxic leachate compounds generates such as Atrazine, Trichloroethylene etc. These contaminants enter the atmosphere and pollute the main components of environment such as atmosphere, lithosphere (land) and hydrosphere (water). The generated toxic leachate also percolates through the underlying soil layer. These contaminants contain extensive ranges of possible carcinogens, non-carcinogens and toxicological compounds that signify a potential risk to the public health. So, it has become important to know the possible concentration, mass and percentage of these chemicals in different waste components like landfill gas, leachate and waste (soil) to extract the behaviour profile of these contaminants in the environment. In this study, an attempt is made to find the behaviour of three priority contaminants in landfill waste on the basis of Level II Fugacity model. The behaviour here actually means concentration, mass distribution, fugacity, rate of reaction, rate of advection etc. of selected contaminants which releases from the waste landfill site at old Rajbandh.

1.2 Rationale

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 D. et al., 1985). 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.

The Level II 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) and octanol water partition coefficients (K_{ow}) need to be specified.

Using the input parameters, Henry's Law Constant (H) is determined using the following Equation 1,

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

Where, H is the Henry's Law Constant ($Pa^3 m/mol$), P_s is Vapor pressure (Pa) and C_s is Aqueous solubility (mol/m^3)

Oxidation, hydrolysis, photolysis and biodegradation reaction were considered in each of the compartment for the decay or removal of the contaminant from the evaluative environment. The combined values of first order reaction rate constant were obtained from Equation 2.

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

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})

The fugacity capacities (Z) of contaminant at different compartment of the evaluative environment are obtained using the formulas presented in Table 1.

Table 1: Definition of fugacity capacities

Compartment	**Fugacity capacity, Z(mol/m³Pa)**	**Source of parameters**
LFG/air (1)	$1/RT$ [a]	R = 8.314 Pa³/mol K T= Absolute temperature (298 K)
Leachate (2)	$1/H$ or C_S/P_S [b]	H = Henry's Law Constant (Pa³m/mol) C^S = Aqueous Solubility (mol/m³) 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 x K_{ow} Where, x = Fraction of organic carbon K_{ow} = Octanol-water partition coefficient

[a, b, c] (Mackay D. et al., 1985)

The contaminant can be removed from different compartment of the evaluative environment by advection and reaction. To acknowledge advection and reaction, a term called D value arises which is further used for calculating the rate of advection and reaction separately. D values for advection can be found using Equation 3.

$$D_A = GZ \tag{3}$$

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.

$$D_R = VKZ \tag{4}$$

Where, D_R denotes D values for reaction (mol/Pa h), V is volume of the compartment (m³), K is combined reaction rate constant (h^{-1}) and Z is fugacity capacity (mol/m³ Pa)
Mass balance equation for each compartment yields a general form like Equation 5.

$$E_i = D_A f + D_R f \tag{5}$$

Where, E_i is emission rate of contaminant at compartment *i* (mol/h), D_A is D value for advection (mol/Pa h), D_R is D values for reaction (mol/Pa h) and f is fugacity of the contaminant at compartment *i* (Pa).

Lumping Eq. (5) 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. The concentration, rate of advection and rate of reaction of contaminants can be obtained from Equations 6, 7 and 8, respectively.

$$C = fZ \tag{6}$$
$$E_A = D_A f \tag{7}$$
$$E_R = D_R f \tag{8}$$

Where, C denotes Concentration (mol/m³), E_A denotes rate of advection (mol/h) and E_R denotes rate of reaction (mol/h). Equations (6), (7) and (8) represents the behaviour of contaminants in LFG, leachate and waste compartment, respectively, of the evaluative landfill envirionment.

1.3 Major Contaminants in landfill waste

In the process of MSW degradation in landfills, 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 carcinogenous and non-carcinogenous behaviour. This study focussed on atrazine, trichloroethylene and m-xylene as representative of main types of priority contaminants.

1.4 Study aims

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 II Fugacity model. The objectives of this study were to (i) characterize the fate of these contaminants in the evaluative environment based on 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.

2. MATERIALS AND METHODS

The methodology adopted in this study is highlighted and hence discussed in the following articles.

2.1 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 and characteristics of the evaluative environment are reported in Table 2. The volumetric composition and physical characteristics of the environment (Table 3) provide a valuable base for this study.

Table 2: Dimension of evaluative environment

Parameter	Value
Cell area	5.42E+04 m^2
Cell depth	4 m[a]
Cell volume	2.17E+05 m^3
Waste density	1.00E+03 kg/m^3[g]
Waste deposition rate	260 ton/day[h]

Table 3: Volumetric composition of environment

Parameter	Volume fraction	Volume
LFG/air	0.10[d]	2.17E+04 m^3
Leachate	0.65[e]	1.27E+05 m^3
Waste	0.25[f]	6.83E+04 m^3

[d, e, f, g, h] (Islam M.R., 2014)
[h] (Khan M.S. et al., 2015)

2.2 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 D. 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 D. et al., 1985) were used as the chemical input parameters such as emission rates, reaction rate constants, fraction of organic carbon, etc. (Table 4 and 5).

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 (MCS) using @RISK 7.6, these chemical input parameters were varied in nature of a lognormal distribution curve with suitable mean and standard deviation values supporting the literatures.

Table 4: Chemical properties and emission rates

Chemical Name	**MW (g/mol)**	**C^s (g/m^3)**	**P^s (Pa)**	**$LogK_{ow}$**	**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

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

2.3 Fugacity modelling

Level II fugacity calculations illustrate the partitioning behaviour of contaminants in the evaluative environment of landfill. Level II model accounts for equilibrium, steady state and flow system i.e. the amount of contaminant entering the environment is mass balanced by the amount lost to flow, reaction or degradation (Shafi S. et al., 2006). The entire phenomenon is briefly illustrated in Figure 1 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.

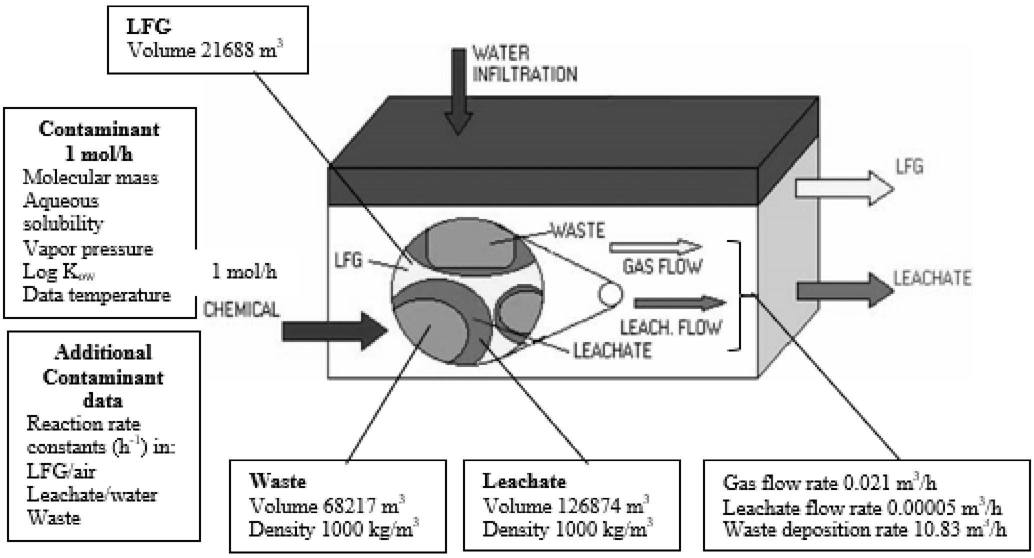


Figure 1: Evaluative environment for Level II Fugacity model

The characteristics of the system are presented in Table 6. Using Equations (1) to (8), the behaviour such as concentration, fugacity, mass distribution, rate of advection, rate of reaction etc. of specific contaminants in LFG, leachate and waste compartment were characterised.

Table 6: Parameters for Level II Fugacity model calculation

Parameter	value	Justification/Reference
Emission rate into environment	1 mol/h	Taken from (Mackay D. et al., 1985)
Landfill gas flow rate	2.10E-02 m^3/h	Adapted to evaluative environment [i]
Leachate flow rate	5.00E-05 m^3/h	Adapted to evaluative environment [j]

[i, j] (Islam M.R., 2014)

2.4 Monte Carlo simulation

As models are only approximation of the contaminant's actual behaviour in real environment, communicating the uncertainties associated with the model is crucial (Kilic S.G. & Aral M.M., 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 (Kumar M.P., 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 and contaminant emission rates in different compartments have been assigned lognormal distribution as it has a positive state space. 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 5000 trials were performed using @RISK 7.6 for the Monte Carlo runs with 5000 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 II Fugacity model outcomes and the most sensitive parameters were recognized from tornado charts.

3. RESULTS AND DISCUSSION

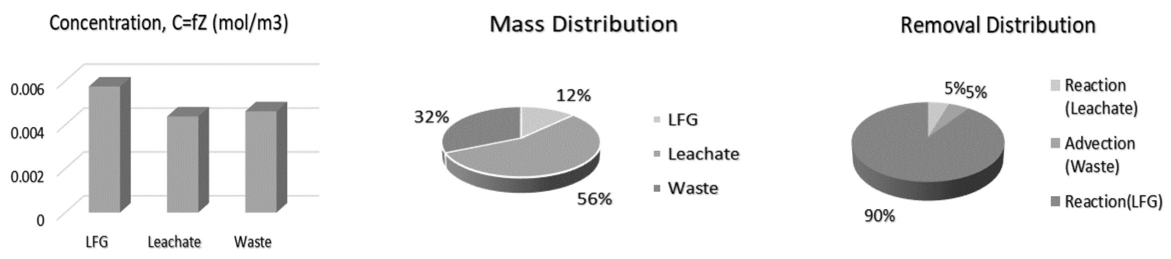
The findings from Level II Fugacity model and Monte Carlo simulation are presented and hence discussed in the following articles.

3.1 Level II Fugacity Modelling

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

3.1.1 Trichloroethylene

The modelled concentration of Trichloroethylene was found 5.70E-03 mol/m^3, 4.40E-03 mol/m^3 and 4.60E-03 mol/m^3 in LFG, leachate and waste compartment, respectively (Figure 2a). High concentration was found in the LFG compartment (Figure 2a) because Trichloroethylene has high volatility and high emission rate in LFG media. Most of the mass was found in leachate compartment (Figure 2b) because of its high volume. The behaviour depicted in Figure 2c showed that about 90% of the total emitted Trichloroethylene was removed by reaction (oxidation) in LFG, 5% by advection in waste and the remaining (5%) was removed by reaction (hydrolysis) in landfill leachate. About 95% of total emitted amount was removed by reaction. So, advection was relatively unimportant compared to reaction as a removal mechanism for Trichloroethylene.

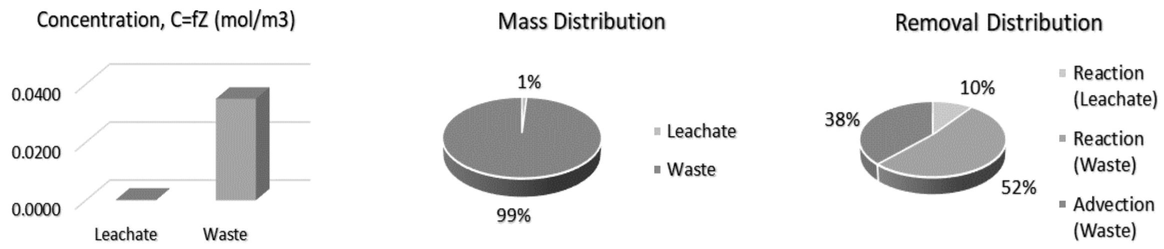


(a) (b) (c)

Figure 2: Behaviour of Trichloroethylene in evaluative environment

3.1.2 Atrazine

The concentration of Atrazine was found approximately 2.00E-04 mol/m^3 and 3.50E-02 mol/m^3 in leachate and waste compartment, respectively (Figure 3a). No mass and concentrations of atrazine was found in LFG compartment as there was no emission of it in LFG and no intermedia transfer was considered. About 98.93% of total mass was found in leachate and remaining 1.07% was in waste (Figure 3b). The illustration in Figure 3c showed that about 52% Atrazine was removed by reaction (photolysis and hydrolysis) in waste, 38% by advection in waste and the remaining by reaction (biodegradation) in leachate. Reaction (photolysis, hydrolysis and biodegradation) was the main removal mechanism for Atrazine as about 62% of the total emitted amount was removed by reaction.

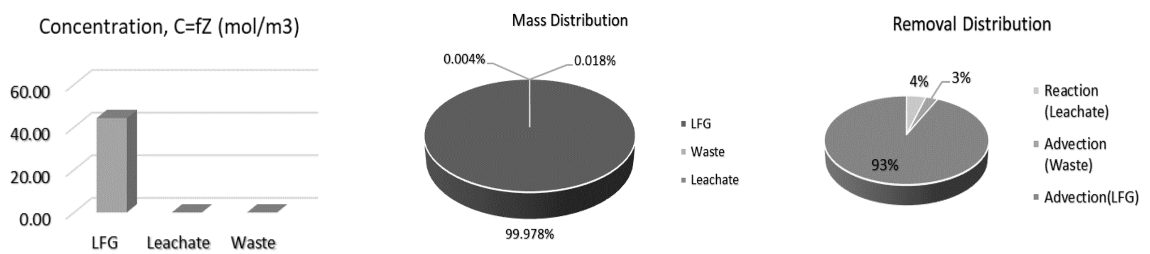


(a) (b) (c)

Figure 3: Behaviour of Atrazine in evaluative environment

3.1.3 m-Xylene

The concentration of m-Xylene was found 44.29 mol/m^3, 3.30E-04 mol/m^3 and 2.50E-03 mol/m^3 in LFG, leachate and waste compartment, respectively (Figure 4a). High concentration and most of the mass (Figure 4b) was found in the LFG compartment because of its high emission rate in LFG media, high volatility and relatively low aqueous solubility. Figure 4c showed that about 93% of the total emitted m-Xylene was removed by advection in LFG, about 4.3% by reaction in leachate and the remaining 2.7% by advection in waste. So, advection was the main removal mechanism for m-Xylene. Though m-Xylene has very low concentration in leachate, it was the only compartment where reaction (oxidation and biodegradation) could occur for this contaminant. This is why a little portion (4.3%) of the total emitted m-Xylene was removed by reaction in leachate.



(a) (b) (c)

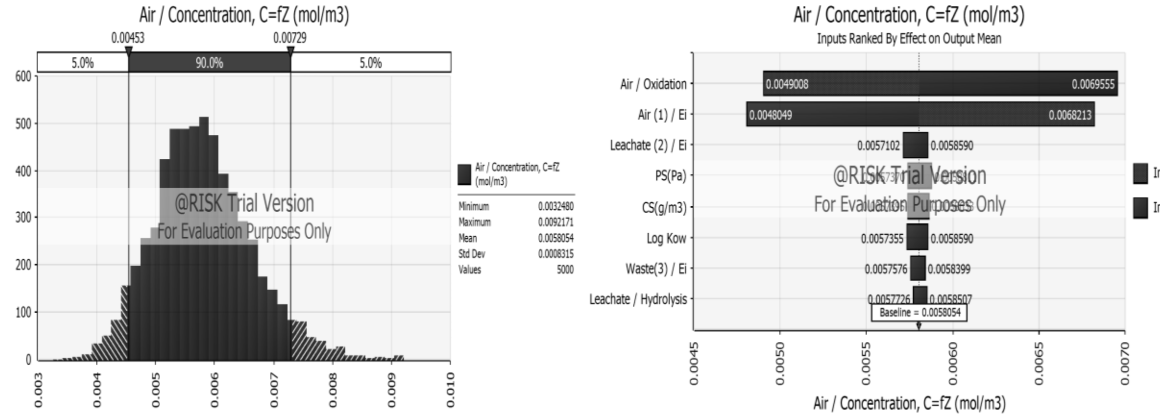
Figure 4: Behaviour of m-Xylene in evaluative environment

3.2 Monte Carlo simulation

To fit a lognormal distribution of the input parameters, this highest concentration was compiled using @RISK 7.6. The MCS was run for 5000 trials with 5000 random variables for the input parameters of Fugacity model. The most sensitive parameter in Monte Carlo runs was found by varying each of the input parameter within the assigned lognormal distribution curve and keeping the other parameters at their static values.

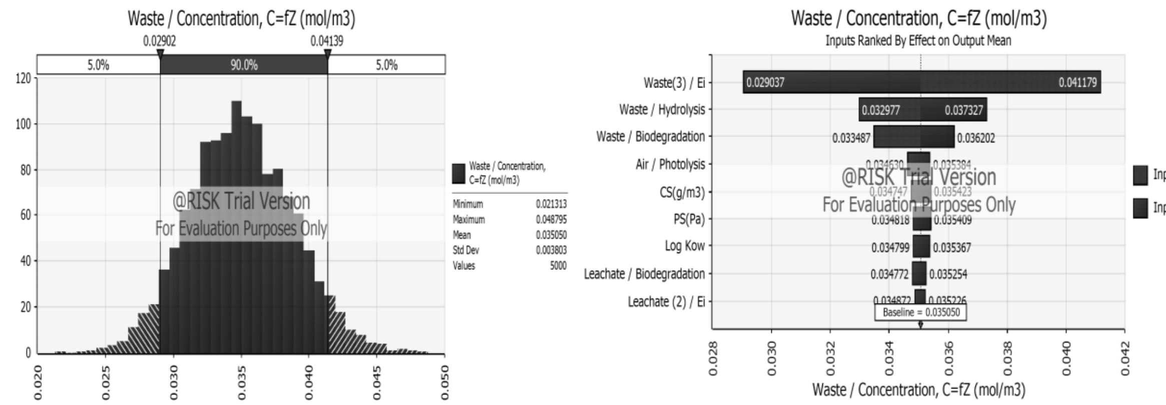
3.2.1 Trichloroethylene

Highest concentration of Trichloroethylene was found in LFG compartment with a value of 5.70E-03 mol/m^3. In Figure 5a, 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 5a, it is seen that the concentration of Trichloroethylene in LFG media ranges from 3.00E-03 mol/m^3 to 4.53E-03 mol/m^3 and 7.29E-03 mol/m^3 to 1.00E-02 mol/m^3 for the 5^{th} and 95^{th} percentile, respectively. The concentration of 4.53E-03 mol/m^3 responds to 5^{th} percentile and 7.29E-03 mol/m^3 responds to 95^{th} percentile. So approximately, 90% (i.e. 0.95-0.05 = 0.90) of the concentration is likely to be exist between 4.53E-03 mol/m^3 to 7.29E-03 mol/m^3 with a mean value of 5.80E-03 mol/m^3. The modelled concentration was also found within this range. It was also found that oxidation reaction rate constant and emission rate in LFG compartment was the most sensitive parameter for variation of this concentration in Monte Carlo runs (Figure 5b). The concentration increases if the oxidation reaction rate constant in LFG compartment decreases and vice versa (Figure 5b). It also increases if the emission rate of Trichloroethylene in LFG media increases and vice versa.



(a) (b)

Figure 5: Monte Carlo simulation run for Trichloroethylene



(a) (b)

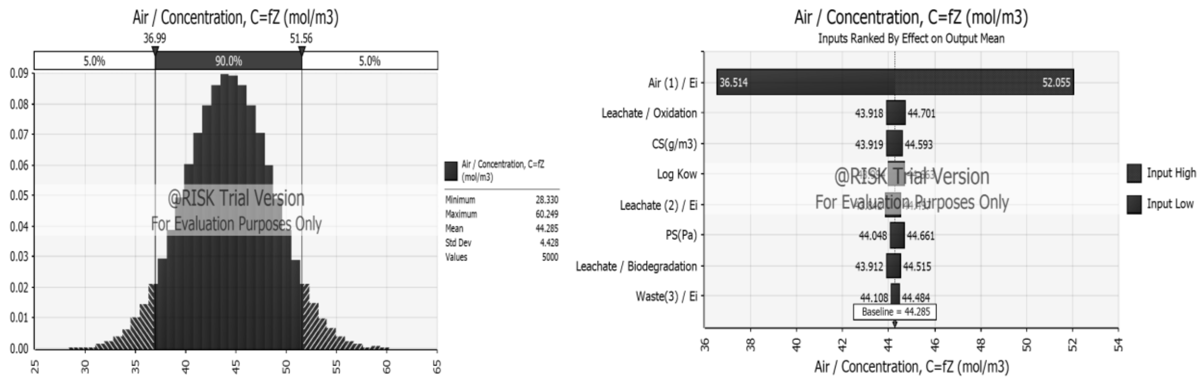
Figure 6: Monte Carlo simulation run for Atrazine

3.2.2 Atrazine

Atrazine had highest concentration in waste compartment with a value of 3.50E-02 mol/m^3. In Figure 6a, 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 6a, it is seen that the concentration of Atrazine in waste media ranges from 2.00E-02 mol/m^3 to 2.90E-02 mol/m^3 and 4.14E-02 mol/m^3 to 5.00E-02 mol/m^3 for the 5^{th} and 95^{th} percentile, respectively. The concentration of 2.90E-02 mol/m^3 responds to 5^{th} percentile and 4.14E-02 mol/m^3 responds to 95^{th} percentile. So approximately, 90% (i.e. 0.95-0.05 = 0.90) of the concentration is likely to be exist between 2.90E-02 mol/m^3 to 4.14E-02 mol/m^3 with a mean value of 3.51E-02 mol/m^3. The modelled concentration was also found within this range. The most sensitive parameter for the variation of this concentration in Monte Carlo runs was emission rate in waste media (Figure 6b). The concentration increases if the emission rate of Atrazine in waste media increases and vice versa.

3.2.3 m-Xylene

m-Xylene had highest concentration in LFG compartment with a value of 44.29 mol/m^3. In Figure 7a, 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 7a, it is seen that the concentration of m-Xylene in LFG media ranges from 25 mol/m^3 to 36.99 mol/m^3 and 51.56 mol/m^3 to 65 mol/m^3 for the 5^{th} and 95^{th} percentile respectively. 36.99 mol/m^3 responds to 5^{th} percentile and 51.56 mol/m^3 responds to 95^{th} percentile. So approximately, 90% (i.e. 0.95-0.05 = 0.90) of the concentration is likely to be exist between 36.99 mol/m^3 to 51.56 mol/m^3 with a mean value of 44.285 mol/m^3. The modelled concentration was also found within this range. Most sensitive parameter for the variation of this concentration in Monte Carlo runs was emission rate in LFG media (Figure 7b). The concentration increases if the emission rate of m-Xylene in LFG media increases and vice versa.



(a) (b)

Figure 7: Monte Carlo simulation run for m-Xylene

3.3 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 D. et al., 1985) were considered including emission rate, aqueous solubility, vapor pressure, octanol-water partition coefficient and so on. 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 further limitation is evaluating the model at standard temperature of 25℃ though landfill will undergo diurnal, seasonal and microbiologically induced temperature changes through various stages of its lifetime (Shafi S. 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. A further and significant limitation was choosing the Level II Fugacity approach rather than Level III and IV approach. Level II Fugacity approach doesn't address the intermedia transport of the contaminants between the compartment of the evaluative environment. Due to lack of intermedia transport coefficient data, Level II approach was chosen and it was not capable of representing the actual complex behaviours that contaminants were undergoing in the landfill environment.

4. CONCLUSIONS

Implementing Level II Fugacity model on complex landfill environment like old Rajbandh have shown how contaminants like Trichloroethylene, Atrazine and m-Xylene partitions among the landfill media in a simplified way. Trichloroethylene and m-Xylene had the highest concentration in landfill gas, while, Atrazine had the highest concentration in waste. The model also showed how these contaminants could be lost or removed from the landfill environment without considering intermedia transportation of the contaminants. Atrazine and Trichloroethylene was mostly removed by reaction, while, advection was the main removal mechanism for m-Xylene. Monte Carlo simulation was introduced here to reduce the variability and uncertainty associated with the input parameters addressed in the model. As standard data set regarding the model outcomes for the selected landfill site was sparse, Monte Carlo simulation justified the model outcomes primarily. There is an explicit need for precise site-specific chemical input parameters and emission rates to increase the efficiency of this model. More accurate outcomes would have been found if intermedia transportation of contaminants were taken into account. Landfill pose health hazard to the local communities and workers. To address health hazard, it is a must to know the behaviour of potential harmful contaminants. Notwithstanding several limitations, this study had illustrated the application of Fugacity model on determining the potential behaviour of contaminants generated from landfill.

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