

# **Development of a Fuzzy-Set Enhanced Environmental Multimedia Modelling System**

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A Thesis  
in  
The Department  
of  
Building, Civil and Environmental Engineering

Presented in Partial Fulfillment of the Requirements  
for the Degree of Master of Applied Science (Civil Engineering) at  
Concordia University  
Montreal, Quebec, Canada

August 2006

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# **ABSTRACT**

## **Development of a Fuzzy-Set Enhanced Environmental Multimedia Modelling System**

Rong Rong Zhang

Environmental pollution problems are crucial concerns that have attracted a great deal of public attention. More actions are urgently required on formulating remediation strategy for contaminated sites and setting effective standard and regulations for reducing the toxic pollutants in the environment. To achieve these needs, comprehensive understanding of and characterizing the natural behaviour of chemical in the environment are essential tasks for environmental risk assessment and management.

In this study, a new user-friendly fuzzy-set enhanced environmental multimedia modelling system (EMMS) is developed, which includes four key modules: an air dispersion module, a polluting source module, an unsaturated zone module, and a groundwater module. Many improvements over previous environmental multimedia models have been achieved, through dynamically quantifying the intermedia mass flux; incorporating fuzzy-set approach into EMMS; and designing a user-friendly graphic user interface (GUI). The developed EMMS can be a useful tool in (1) estimating the time-varying and spatial-varying chemical concentrations in air, soil, and groundwater; (2) characterizing the potential risk to human health presented by contaminants released from a contaminated site; and (3) quantifying the uncertainties associated with modelling

systems and subsequently providing robustness and flexibility for the remediation-related decision making.

The developed EMMS has been tested and validated through a set of case studies including both pilot-scale and field-scale validations. Reasonable results have been obtained. It indicates that, with the aid of fuzzy-set approach, EMMS can be a reliable and powerful tool to address the complex environmental multimedia pollution problems and provide an extensive support for decision makers in managing the contaminated environmental systems.

## **ACKNOWLEDGEMENT**

I would like to express my sincere appreciation to my supervisor, Dr. Zhi Chen, for his guidance, generous suggestion and continuous support during my whole Master studies. Dr. Chen is always there to listen and give timely advice with much patient and encouragement. Not only does he teach me how to approach a research problem with persistence, but also his working philosophy shows me a good example for how to accomplish the goals in the life and in the studies. His guidance, advice, and support are greatly helpful for my future career and I will also benefit enormously from these invaluable assets in my entire life.

Sincere thanks also go to my friends in our group, for their valuable and helpful assistance in any aspects of my research and studies. The lasting friendship between us gives me sufficient encouragement and more inspiration.

I extend my deep gratitude to my father, my mother, and my sister for their unconditional mortal support and never-ending solitude. Special thanks are given to my husband, Zhong Peng He. Without his support and standing by me all the time, I cannot overcome the difficult time I encountered during the whole studies.

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## List of Symbols

### Symbols used in the modules of EMMS

$a$ :	Volumetric air content [dimensionless]
$a_I$ and $b_I$ :	Constant
$A_x$ :	Length of landfill area, parallel to groundwater flow [L]
$A_y$ :	Width of landfill area, orthogonal to groundwater flow [L]
$A(x, y, t)$ :	General form of analytical solution where concentration is a function of space and time.
$B_I$ :	Retardation factor [dimensionless]
$B$ :	thickness of the saturated zone [L]
$B'$ :	Defined in Equation (5.8)
BCF:	Bioconcentration factor for aquatic life [ $L^3 M^{-1}$ ]
$C_{s1}$ :	Solute concentration at boundary after $t=t_1$ in groundwater [ $M L^{-3}$ ]
$C$ :	Concentration in landfill [ $M L^{-3}$ ]
$C_{un}$ :	Concentration in unsaturated zone [ $M L^{-3}$ ]
$C_s$ :	Concentration in situated zone [ $M L^{-3}$ ]
$C_{air}$ :	Concentration in air [ $M L^{-3}$ ]
$C_{L0}$ :	Background concentration in landfill [ $M L^{-3}$ ]
$C_{s0}$ :	Time-weighted average contaminant concentration on the mixing zone [ $M L^{-3}$ ]
$C_0$ :	An initial concentration in landfill, used in case study [ $M L^{-3}$ ]
$C_G$ :	Concentration in gaseous phase [ $M L^{-3}$ ]
$C_L$ :	Concentration in dissolved phase [ $M L^{-3}$ ]
$C_s$ :	Absorbed concentration in soil in landfill [ $M M^{-1}$ ]
$C_T$ :	Total concentration [ $M L^{-3}$ ]

$D_g^a$ :	Gaseous diffusion coefficients in air [ $L^2 T^{-1}$ ]
$D_g$ :	Gaseous diffusion coefficients in soil [ $L^2 T^{-1}$ ]
$D_l^w$ :	Liquid diffusion coefficients in pure water [ $L^2 T^{-1}$ ]
$D_l$ :	Liquid diffusion coefficients in soil [ $L^2 T^{-1}$ ]
$D_L$ :	Coefficient of longitudinal dispersion of unsaturated zone [ $L^2 T^{-1}$ ];
$D_T$ :	Coefficient of transverse dispersion of unsaturated zone [ $L^2 T^{-1}$ ]
$D_x$ :	Dispersion coefficient in x coordinate direction in groundwater [ $L^2 T^{-1}$ ]
$D_y$ :	Dispersion coefficient in y coordinate direction in groundwater [ $L^2 T^{-1}$ ]
$D_z$ :	Dispersion coefficient in z coordinate direction in groundwater [ $L^2 T^{-1}$ ]
$E_i$ :	Chemical inputs to compartment $i$ by direct emission [ $M L^{-2} T^{-1}$ ]
$f_c(\Phi)$ :	Frequency or probability of occurrence of a specific wind speed [dimensionless]
$f(\Phi)$ :	Frequency of wind in direction of exposure location [dimensionless]
$f_{oc}$ :	Organic carbon fraction in soil or waste [dimensionless]
$f_{ocun}$ :	Organic carbon fraction in unsaturated zone [dimensionless]
$f_{ocsat}$ :	Organic carbon fraction in saturated zone [dimensionless]
$F$ :	Fugacity [ $M T^{-2} L^{-1}$ ]
$h$ :	Overall mass transfer coefficient through top cover [ $L T^{-1}$ ]
$H_E$ :	Defined in Equation (3.21) [ $L T^{-1}$ ]
$H$ :	Aquifer mixing zone thickness [ $L$ ]
$J_a$ :	Aqueous and gaseous phase advective flux [ $M L^{-2} T^{-1}$ ]
$J_g$ :	Diffusive vapour flux [ $M L^{-2} T^{-1}$ ]
$J_l$ :	Diffusive flux of dissolved solute [ $M L^{-2} T^{-1}$ ]

$J_t$ :	Total of diffusive and advective fluxes in gaseous and dissolved phase released from landfill [ $M L^{-2} T^{-1}$ ]
$J_{run}$ :	Total of diffusive and advective fluxes of dissolved phase released from soil [ $M L^{-2} T^{-1}$ ]
$k$ :	Fitting parameters in calculation of gas generation potential [ $T^{-1}$ ]
$k_i$ :	Mass transfer coefficient in the air-soil boundary layer [ $L T^{-1}$ ]
$K_{I2}$ :	Partition coefficient [dimensionless]
$K_D$ :	The liquid-soil adsorption coefficient in landfill [ $L^3 M^{-1}$ ]
$K_{D1}$ :	The liquid-soil adsorption coefficient in unsaturated zone [ $L^3 M^{-1}$ ]
$K_G$ :	The gas-soil adsorption coefficient [ $L^3 M^{-1}$ ]
$K_H$ :	Henry's law constant [dimensionless]
$K_{oc}$ :	Equilibrium partition coefficient or organic carbon partition coefficient [ $L^3 M^{-1}$ ]
$K_{ow}$ :	Octanol-water partition coefficient [dimensionless]
$L$ :	Depth of landfill [L]
$L_c$ :	Characteristic length [L]
$m_0$ :	Rate of release of solute from point source at zero time [ $M T^{-1}$ ]
$M_A$ :	Molecular weight of air [ $M M^{-1}$ ]
$M_B$ :	Molecular weight of the compound of interest [ $M M^{-1}$ ]
$M_g$ :	Cumulative gas phase diffusive flux [ $M L^{-2}$ ]
$M_l$ :	Cumulative liquid phase diffusive flux [ $M L^{-2}$ ]
$M_a$ :	Cumulative gas and liquid phase advective flux [ $M L^{-2}$ ]
$M_t$ :	Total cumulative flux in leachate from landfill [ $M L^{-2}$ ]
$M_{run}$ :	Total cumulative flux in leachate from soil [ $M L^{-2}$ ]



$M$ :	Total mass of contaminant released from landfill to unsaturated zone at time $t$ [M]
$M_{vol}$ :	The cumulative mass lost from the landfill surface due to volatilisation [M L <sup>-2</sup> ]
$Q$ :	Concentration of solute in solid phase in unsaturated zone [M L <sup>-3</sup> ]
$Q(t_{ad})$ :	The mass emission rate from the surface of the site to the atmosphere over the annual duration [M T <sup>-1</sup> ]
$Q_f$ :	Percolation rate [L T <sup>-1</sup> ]
$r$ :	Reduced coordinate defined in Equation (3.40) [L]
$R_G$ :	The equilibrium distribution coefficient between total and gaseous phase contaminant concentrations [dimensionless]
$R_L$ :	The equilibrium distribution coefficient between total and aqueous phase contaminant concentrations [dimensionless]
$R_S$ :	The equilibrium distribution coefficient between total and solid phase contaminant concentrations [dimensionless]
$R$ :	Defined in Equation 3.47 [dimensionless]
$S$ :	Gas generation potential [L <sup>3</sup> ]
$S_0$ :	Initial gas generation potential [L <sup>3</sup> ]
$S_o$ :	Chemical- water solubility [M L <sup>-3</sup> ]
$t_{ad}$ :	The total exposure duration [T]
$TR_u$ :	Transfer function in unsaturated zone [L <sup>-2</sup> ]
$TR_s$ :	Transfer function in saturated zone [dimensionless]
$TR_a$ :	Transfer function in air [dimensionless]
$u$ :	Annual average surface wind speed [L T <sup>-1</sup> ]
$U_i$ :	Mass-transfer coefficients in the boundary layers in compartment $i$ [L T <sup>-1</sup> ]
$v$ :	Average velocity of fluid in unsaturated zone [L T <sup>-1</sup> ]
$v'_G$ :	Gaseous interstitial velocity [L T <sup>-1</sup> ]

$v_G$ :	Bulk gas velocity [ $L T^{-1}$ ]
$v_L'$ :	Leachate interstitial velocity [ $L T^{-1}$ ]
$v_L$ :	Bulk water velocity [ $L T^{-1}$ ]
$V_d$ :	Uniform seepage velocity in the groundwater flow (x) direction [ $L T^{-1}$ ]
$V_B'$ :	LeBas molar volume of the compound of concern [ $L^3 M^{-1}$ ]
$Y_{12}$ :	Fugacity mass-transfer coefficient across the boundary between compartments [ $M L^{-2} (M L^{-1} T^{-2})^{-1} T^{-1}$ ]
$Z$ :	Fugacity capacity [ $M L^{-3} (M L^{-1} T^{-2})^{-1}$ ]
$z_{wt}$ :	Depth of water table below the landfill [L]
$\sum flow$ :	Rate of inter-compartmental transfer [ $M L^{-2} T^{-1}$ ]
$\theta$ :	Volumetric water content [dimensionless]
$\rho_b$ :	Bulk density [ $M L^{-3}$ ]
$\rho_{unsat}$ :	Bulk density of saturated zone [ $M L^{-3}$ ]
$\rho_{sat}$ :	Bulk density of saturated zone [ $M L^{-3}$ ]
$\phi$ :	Total porosity of landfill [dimensionless]
$\phi_{un}$ :	Effective porosity of unsaturated zone [dimensionless]
$\phi_s$ :	Effective porosity of saturated zone [dimensionless]
$\mu_{sat}$ :	First-order degradation rate in saturated zone [ $T^{-1}$ ]
$\mu_{unsat}$ :	First-order degradation rate in saturated zone [ $T^{-1}$ ]
$\mu$ :	Effective first order reaction rate constant in landfill [ $T^{-1}$ ]
$\tau$ :	Effective half-life in landfill chamber [T]
$\tau_{unsat}$ :	Half-life in unsaturated zone [T]
$\tau_{sat}$ :	Half-life in saturated zone [T]
$V_E$ :	Effective solute velocity ( $L T^{-1}$ )

$\alpha$ :	Rate of solid volume to fluid volume in unsaturated zone [dimensionless]
$\alpha_x, \alpha_L$ :	The dispersivity in the x coordinate directions [L]
$\alpha_y, \alpha_T$ :	The dispersivity in the y coordinate directions [L]
$\alpha_z, \alpha_V$ :	The dispersivity in the z coordinate directions [L]
$\gamma$ :	Time constant for decay of release rate [ $T^{-1}$ ]
$\eta$ :	Reduced coordinate defined in Equation (3.40) [ $T^{1/2}$ ]
$\eta_w$ :	Viscosity of water [ $M L^{-1} T^{-1}$ ]
$\sigma_z$ :	Vertical dispersion coefficient [L]
$\sigma_{zD}$ :	Vertical dispersion coefficient for D stability class [L]
$\sigma_{AB}$ :	Defined in Equation [5.9]
$\Omega$ :	Defined in Equation [5.10]

### **Symbols used in the fuzzy-set based equations**

a:	Left spread of a fuzzy set
A:	a fuzzy set or an uncertain value
b:	right spread of a fuzzy set
m:	the mean value of a fuzzy set
k:	an ordinary number
M, N:	Fuzzy numbers
R:	a set of real numbers
x:	the objects
X:	a collection of objects
$\mu_A(x)$ :	The membership function or grade of membership

## List of Abbreviation and Acronyms

3MRA:	The Multimedia, Multipathway, and Multireceptor Risk Assessment modelling system
ANL:	Argonne National Laboratory
BETR North America:	A multimedia contaminant fate model for North America
BET:	Benzene, ethylbenzene, and toluene
ChemCAN:	Multimedia model of chemical of fate in Canada
Chamrange:	Multimedia transport model for calculating persistence and spatial range of organic chemicals
CHEMFRANCE:	Regional level III fugacity model applied to France
CalTOX:	Multimedia total exposure model
DOE:	The U.S. Department of Energy
EMMS:	An Environmental Multimedia Modelling System
GUI:	Graphic User Interface
ISMCM:	Integrated Spatial-Multimedia-Compartmental Models
LSSMM:	Linked spatial single-media models
MCM:	the Multimedia Compartment Model
MEPAS:	The Multimedia Environmental Pollutant Assessment System
MMSOILS:	The Multimedia Contaminant Fate, Transport, and Exposure Model
RESRAD:	Computer program for RESidual RADioactivity
SimpleBox:	Multimedia mass balance model
SMCM:	the Spatial Multimedia Compartment Model
USEPA:	The U.S. Environmental Protection Agency
ODWS:	Ontario Drinking Water Standard
OSW:	Office of Solid Waste
ORD:	EPA's Office of Research and Development
TF:	Transfer function
TFN:	Triangular fuzzy number

WMU: Waste management unit  
VOC: Volatile organic compound

# **Chapter 1**

## **Introduction**

### **1.1 The Background of Environmental Multimedia Modelling Development**

Environmental pollution problems are concerns that have attracted a great deal of public attention. Facing the increasing environmental issues that pose a variety of impacts and risks on public health, more actions are urgently required to create a remediation strategy for contaminated sites, setting stringent standards and regulations for reducing the toxic pollutants in the environment, and even determining the priority of the potential environmental problems (Droppo et al. 1993, Hsieh and Ouimette 1994, USEPA 1996). To achieve these goals, significant efforts have been put forth to develop modelling tools and to make quantitative and qualitative risk assessment. Consequently, a comprehensive understanding and characterisation of the natural behaviour of chemicals in the environment are essential tasks for environmental risk assessment and management.

Before the late-1950s, contaminant fate and exposure assessment was

concentrated on assessing the behaviour of single chemical in only air, only water, or only soil. However, the persistency of certain chemical pollutants in the environment, the migration of pollutants between environmental media - air, water, soils and sediments - and the accumulation of pollutants in these media, a novel approach was required in order to carry out research on extensive characterisations of chemical transport in multiple environmental media over regional, continental, and global scales. Namely, research on environmental risk assessment and management should not focus not only on the chemical transport and transformation in one medium, but also on the contaminant transfer rate between environmental media and geo-hydrological processes at the soil/water/air interface. Responding to these requirements (TECO 1985, McKone and MacLeod 2003), environmental multimedia modelling has evolved.

The earliest rudiment of the multimedia model originated in the 1950's because of the need to assess human exposure to global fallout that requires considering the transport in air, soil, surface water, vegetation and the food chain (Eisenbud 1987, Ng 1982, and Whicker and Kirchner 1987).

In the 1970s, growing concerns about the impacts of metals that were distributed in multiple environmental media, such as lead, cadmium, mercury, and arsenic, bring on the rapid development of global and regional mass-balance models for these metals (Garrels et al. 1975, and Nriagu 1978, 1979). Until the late 1970s, the seminar papers by Mackay describing the application of fugacity principles to environmental problems remarked the regional/global mass-balance models for organic chemicals had clearly

emerged (Mackay 1979, Mackay and Paterson 1981, 1982). By the late 1980s multimedia modelling had become more established (Cohen 1986, Allen et al. 1989, Machay 1991, and Cowan et al. 1995).

Basically, multimedia models can be classified into three types (Hsieh and Ouimette 1994, Cohen and Cotter 2002):

1. Compartmental (“well-mixed” media) models;
2. Integrated spatial-multimedia-compartmental models (ISMCM); and
3. Linked spatial single-media models (LSSMM).

Of these, the most widely used is the compartmental mass-balance model, which has become popular over the last decade due to its simple and easy execution. Particularly, the concept of fugacity in compartmental multimedia models has gained the most attention. The major limitation of this kind of model is the key assumption that all the compartments are uniformly mixed. Such an assumption is obviously not correct when the effect of variation in the spatial and temporal scale of the model is important for the risk assessment of a single point contaminant source, especially, this point source is in soil. Also it is invalid when the scale is small.

ISMCM includes both “well-mixed” and spatial compartments integrated through inter-media physical boundary conditions. The atmosphere and water compartments are treated as uniform multiple compartments. Soil is expressed as a spatial compartment. Thus a more elaborate resolution can be gained in soil, but the approach for air and water compartments still falls short of spatial resolution.



LSSMM integrates together serially linked single-medium transport models that are for air, water, soil and other media of interest (Cohen and Cooter 2002). Compared to other types of multimedia models, it can provide fine spatial and temporal resolutions, which are significant features in evaluating the risk level for exposure to hazardous contaminants.

The principal representatives of LSSMM are MEPAS (Mills et al 1997; McDonald and Gelston 1998); MMSOIL (USEPA 1988); HWIR (USEPA 1999b); MULTIMED 2.0 (Solhotra et al. 1995); and 3MRA(USEPA 2003).

MEPAS and MMSOILS calculate the gas volatilisation from the surface of a polluted site with steady-state empirical equations in scenarios of contaminant release, which is based on the premise of a very large source, so that emission does not deplete the source during the time considered. Obviously, this kind of assumption is far away from the natural behaviour of contaminants in the environment. (USEPA 1996, 2003). 3MRA modified Jury's model (Jury et al.1983, 1990) to estimate a land-based source term; however, it does not consider gaseous advection in its source term estimation, which to some extent affects the strength of the chemical release from the landfill cover and the bottom (Lin and Hildemann 1995).

MEPAS and 3MRA contain a time series of an annual step or a user desired time step, based on which averaged leachate release is calculated. MMSOILS uses a continuously mixed reactor model to simulate a leachate source. These assumptions can be useful in risk evaluation at the screening level, but in reality, a leachate release is not

always in a steady state, but gradually decreases with respect to time.

In this brief introduction of the major environmental multimedia models, it is clear that LSSMM among three kinds of multimedia models is one of the best available choices to fit into the goal of simulating and characterising the behaviour of chemical release from contaminated soil into the environment while providing fine temporal and spatial resolutions.

However, there are limitations in the previous approaches related to LSSMM.

The limitations are as follows:

- 1) Using an empirical algorithm to estimate the gaseous emission from the surface of a contaminated site;
- 2) No consideration of gaseous advection in the polluted soil; and
- 3) Steady state leachate release from the contaminated soil.

Aiming to mitigate the limitations given above and to extend the previous studies on environmental multimedia models, a new environmental multimedia modelling system (EMMS) is developed in this study.

In addition to the consideration of the contaminant behaviour in the environment, a pollutant-releasing source, landfill, has raised other significant concerns during the last two decades. Measurements taken at actual landfill sites of organic chemicals in leachate and gas suggest that emissions of leachate and gas are substantial fate routes (Hogdson et al. 1992, Khim 1994, and Liu et al. 2004). Thus, more and more attentions are focused on the potential emission from landfill, especially, regarding the health risks associated with

trace constituents of these emissions.

In response to this anxiety about the adverse health effects caused by landfills or other contaminated sites, the landfill source term, which can also be used for contaminated sites, is considered as the starting module in the development of EMMS. Three basic environmental media, air, soil and groundwater, are treated as contaminant transport pathways integrated into EMMS for predicting the potential risk at exposure sites. It is expected that this study can promote development in the field of evolving environmental multimedia model, can improve the understanding and knowledge of the methods that describe the chemical transport and transformation in environment, and can lead to the discovery of a better way to deal with the inherent uncertainties of modelling system.

## **1.2 Objective of This Study**

The objective of this study is to develop a user-friendly fuzzy-set enhanced environmental multimedia modelling system (EMMS-PRO), which allows (i) an estimation of the time-varying and spatial-varying chemical concentrations in air, soil, and groundwater, and (ii) subsequently the characterisation of the potential risk to human health presented by contaminants released from a pollution site. The following new improvements over previous LSSMMs will be incorporated into EMMS-PRO:

- 1) A pollutant source model is derived. It is based on the mechanisms of a

pollution site, such as landfill, which represents the complexities associated with the emission into a multimedia environment.

2) The air pollutant emitted from the polluting source is considered with its emission rate and velocity information.

3) The pollutant fluxes from the pollution source into the soil and the groundwater is time-dependant; thus, it is considered as a non-steady-state input source for an unsaturated zone module.

4) The developed EMMS will be enhanced by a fuzzy-set method to quantify a large amount of uncertainties associated with the multimedia environment, the model itself, the model inputs, and the model outputs. It is also intended to provide robustness and flexibility for remediation-related decision making.

### **1.3 Organization of This Thesis**

The thesis is organised as follow:

Chapter 1 gives the background and rationale for this thesis study and also includes the research objectives.

A literature review is presented in Chapter 2 for expanding the knowledge of multimedia modelling development and providing the rationale for developing a new environmental multimedia modelling system.

Chapter 3 describes the derivation of the key modules of EMMS and the

development of the fuzzy-set enhanced EMMS.

Chapter 4 is the first part of the validation of the developed fuzzy-set enhanced EMMS. It includes a testing of the overall performance of EMMS through a case study, and importantly, the comparison with all available literature data and similar previous studies.

Chapter 5 is the second part of system validation, which is conducted using a field case with field historical data.

Chapter 6 contains the development of a graphic user interface for the developed system, which makes it as a user-friendly fuzzy-set enhanced EMMS. The finalized version of the system is applied to the Trail Road landfill in Ottawa-Carleton Region.

Chapter 7 gives conclusions, a summary of research contributions, and recommendations to future study.

## **Chapter 2**

### **Literature Review**

In the past decades, significant attention has been focused on the long-term behaviour and effect of the chemicals released from modern industrial economies into the environment.

In this context, the multimedia-modelling paradigm is becoming an effective and appropriate approach. It can be used to make a complete assessment through accounting for the complex features in the physical, chemical, and even biological contaminant fate and transport processes in the entire environmental system. These approaches consider the locations of the chemical release, the characteristics of inter-media mass distribution, and the pathways of chemical transport; hence, the results from these modelling systems will effectively and efficiently identify where the chain-of-event control effort would be the most operative (McKone and MacLeod 2003).

From the emergence of the multimedia model until now, multimedia modelling approaches can be divided into three types, depending on the level of complexity and the treatment of spatial and temporal resolution:

1. Fugacity-based compartmental model;

2. Integrated spatial-multimedia-compartmental models (ISMCM); and
3. Linked spatial single-media models (LSSMM).

The objective of the present literature review is to survey the previous research efforts conducted in the field of environmental multimedia model evolution. Most representative multimedia models are reviewed in this chapter.

## **2.1 Fugacity-Based Environmental Multimedia Modelling**

### **2.1.1 Model Description**

The property of fugacity makes it easy to use in mass-balance calculations to describe the behaviour of chemicals released into the environment and to simulate their distribution in different environmental media (Mackey 1979, Mackey and Paterson 1981). Hence, this type of multimedia model is widely recognized and widely used.

Fugacity-based model takes into account a variety of transport processes and chemical reactions with the assumption that all the environmental media under consideration are well mixed.

According to the complexity of modelling, fugacity-based environmental fate model is classified into four levels. Level I assumes equilibrium or equi-fugacity among all the compartments of a closed system containing a constant mass of contaminant. Level II provides for flow of the contaminant through the system while remaining the equilibrium between compartments. Level III removes the equilibrium constraint and

allows the mass to in a steady state move in and out of compartments but the overall inventory in each compartment remains constant with time, in other words, steady state modelling. Finally, in Level IV, the constraint of constant mass flux into the system is replaced by an unsteady state, which allows input, and consequently output, to vary with time (Maddelena and McKone 1995, Mackey 1991).

Within the screening framework, Level III is adequate to estimate probable chemical fate with average concentrations in environmental compartments. Thus, most existing fugacity-based multimedia models are built on Level III.

Firstly, the concept of Level III is briefly introduced in this section.

#### Fugacity and Fugacity and Capacity

Fugacity,  $f$ , is a metric for quantifying chemical activity at low concentration (Pa). It can be viewed as the “escaping tendency” of a chemical in a phase, has dimensions of pressure, and is related to concentration,  $C$  (mol/m<sup>3</sup>), by a proportionality constant, fugacity capacity,  $Z$  (mol/m<sup>3</sup>-Pa) (McKone and MacLeod 2003, Mackey 1991).

$$C = Zf \quad [2.1]$$

When a chemical reaches equilibrium distribution between two available phases, the fugacities of the chemical in the phases are equal. Equilibrium partitioning between two phases,  $K_{12}$ , is a dimensionless partition coefficient, and can be measured under laboratory conditions as the ratio of concentrations  $C_1$  and  $C_2$ . Applying the relationship



between concentration and fugacity, and recognizing that  $f_1 = f_2$  at equilibrium:

$$K_{12} = \frac{C_1}{C_2} = \frac{fZ_1}{fZ_2} = \frac{Z_1}{Z_2} \quad [2.2]$$

Z can thus be determined experimentally for many phases by measuring partition coefficients between the phase of interest and a phase with known Z.

#### The Governing Equation of Level III Fugacity Model

The mass-balance condition is applied to each environmental compartment. This requires the quantification of diffusion and advection rates at the compartment boundaries.

The net diffusive flux, in mol/m<sup>2</sup>-h, across the interfacial area separating compartments is:

$$flux = Y_{12}(f_1 - f_2) \quad [2.3]$$

where  $Y_{12}$  is the fugacity mass-transfer coefficient across the boundary between Compartments 1 and 2 with units mol/(m<sup>2</sup>-Pa-h), and  $f_1$  and  $f_2$  are the fugacities of Compartments 1 and 2.

The fugacity mass-transfer coefficient depends on the mass-transfer coefficient on either side of the interface and on the fugacity capacities of the two media that form the interface.

$$Y_{12} = \left( \frac{1}{Z_1 U_1} + \frac{1}{Z_2 U_2} \right)^{-1} \quad [2.4]$$

where  $U_1$  and  $U_2$  are the mass-transfer coefficients (m/h) in the boundary layers in

Compartments 1 and 2, and  $Z_1$  and  $Z_2$  are the fugacity capacities of Compartments 1 and 2.

The inter-compartment transfer of contaminants by advection is modelled as a flux ( $\text{mol/m}^2\text{-h}$ ) at the compartment boundary, which is the product of the velocity of the moving phase ( $\text{m/h}$ ) and the contaminant concentration in that phase ( $\text{mol/m}^3$ ).

$$\text{Advective flux} = \text{velocity} \times Z_{ik} f_i \quad [2.5]$$

where  $Z_{ik}$  and  $f_i$  represent the fugacity capacity of the moving phase and the fugacity of the chemical, respectively, in compartment  $i$ .

Mass balance equations including all inter-compartmental transfers of contaminants can be written for each compartment ( $i$ ) in the environmental system.

$$E_i + \sum \text{flow}_{j \rightarrow i} = \sum \text{flow}_{i \rightarrow j} + \sum \text{flow}_{i \rightarrow \text{sink}} \quad [2.6]$$

On the left-hand side of Equation [2.6] are chemical inputs to compartment  $i$  by direct emission,  $E_i$ , and the total rate of inter-compartmental transfer to compartment  $i$ ,  $\sum \text{flow}_{j \rightarrow i}$ . Removals from the compartment occur by inter-compartmental transfer,  $\sum \text{flow}_{i \rightarrow j}$ , and by advection out of the system or chemical transformation  $\sum \text{flow}_{i \rightarrow \text{sink}}$ . For an environment consisting of  $n$  compartments, one can write  $n$  equations of this type and solve them algebraically to obtain the unknown fugacity of chemical in each compartment. In sequence, the relative concentration of compartments can be computed on the ranking of long-term screening model.

### 2.1.2 Variants of Fugacity Model

ChemCAN was developed at the Canadian Environmental Modelling Centre (CEMC) in 1997 (Mackay et al. 1991, Webster et al. 2004). The new version 6.0 was released in September 2003. ChemCAN is a Level III model of 24 regions of Canada. It estimates average concentrations of contaminants in air, fresh surface water, fish, sediments, soils, vegetation, and marine near-shore waters. It is intended to assist in human exposure assessment. For the modelling application, areas should have a radius of at least 300 km. If investigations involve smaller areas, this model is not suitable.

Similarly, Scheringer et al. (2002) designed ChemRange for the calculation of a chemical's persistence and spatial range in an evaluative model system. It serves as a tool for comparing organic chemicals in terms of their exposure behaviour, and provides a consistent framework for this kind of quantitative chemical comparison and for an exposure-based assessment (Berg and Scheringer 1994, Scheringer 1996,1997). Three compartments, called surface soil, oceanic surface water, and tropospheric air, are linked by diffusive and advective exchange processes to compose the model system. Three compartments include around a circular model world. The concentrations of chemicals in the compartments are the basic model outputs. The emphasis of the model outputs, however, is on the chemical's persistence, spatial range, and mass fractions in the three compartments. Also, the mass flows from three media into four pathways are calculated. Such mass flows include reaction, removal, transfer to the lined media, and deposition to

the deep sea.

The following assumptions and limitations need to be kept in mind when this model is used (Scheringer et al. 2002):

1) The exchange processes between three environmental media are applied only to organic chemicals; the behaviour of metals or ion-forming compounds cannot be investigated with this model;

2) Since the averaged concentration distribution is estimated with this model, the spatial range is a relative measure of the long-range transport potential. The persistence does not describe the actual duration of an exposure but is a relative measure of a chemical's overall degradability;

By comparison, Devillers et al. (1995) developed CHEMFRANCE, a six-compartment multimedia model, for use particularly in France. Ten sub-compartments are embodied in the bulk compartments for the purpose of comprehensive considerations of the media having effects on the transport and transformation of pollutants. The model is constructed on the basis of the fugacity concept and requires information related to the chemical property and emission rates of pollutants. The outputs of the model consist of the chemical distribution between the environmental media, the transport and transformation process rate, and the steady state concentration in the defined regions of France, or in France as a whole.

A case study was performed to illustrate the model application. The simulation results are compared with the observed concentration in the air as a validation test. The

concentrations in locations relatively far from the sources of emission are in accordance with the modelling results, but those near the emission sources are significantly higher. From these studies, it appears that this model is particularly suitable as a regional multimedia model for estimating the environmental fate of organic chemicals in France.

Fourthly, a representative, widely used eight-compartment regional and dynamic multimedia fugacity model, CalTOX, was developed by Environmental Scientists at the University of California campuses and national laboratories (McKone and Enoch 2002). Like Mackey's fugacity model, it calculates a mass balance between gain and loss for each compartment in the system.

Mainly in one way, however, it differs from Mackey's model (Maddelena and McKone 1995). CalTOX calculates gain and loss between compartments as a product of a first order rate constant and the molar inventory, rather than using the product of a fugacity-based transfer coefficient and the source compartment fugacity. Even though this method is essentially same as that of Mackey's model, the use of molar inventory as a state variable makes it easier for CalTOX to evaluate the environmental fate of compounds, such as heavy metals or radionuclides, when a measurable vapour pressure is not available.

By contrast, BETR North America is a regionally segmented multi-compartment, continental-scale, mass balance chemical fate model. Twenty-four ecological regions in North America's environment, divided by geographic features such as waterways and soil types, are modelled (MacLeod et al. 2001).

A system of seven compartments, namely upper atmosphere, lower atmosphere, vegetation, soil, fresh water, coastal water, and freshwater sediment, is modelled for each region on the basis of the fugacity concept. Seven equations describe the contaminant fate in each region of the model, so 168 mass balance equations are needed to constitute the model for the 24 regions of North America.

Furthermore, BETR North America is the first approach to model the movement of persistent organic pollutants on a continental scale, not like most existing multimedia models, such as ChemCAN, CalTOX, and Simple Box, which describe the fate and transport of contaminants only on a smaller regional scale. The BETR North America model can be a valuable tool for assessing the long-range transport potential of persistent organic pollutants and other contaminants.

Finally, the reviewed model in this study is SimpleBOX that is developed and updated by den Hollander and van de Meent (2004). It consists of nine well-mixed homogeneous compartments, namely air, fresh water, seawater, sediments, three soil compartments, and two vegetation compartments, in regional, continental and global scales. It can be implemented as a steady-state, non-equilibrium partitioning, mass balance model. It can also produce non-steady-state output by using an external numerical integrator. Not only is SimpleBox a generic model, but it can also be customised to represent specific environmental situations.

Unlike the fugacity approach adopted by Mackey, computation of mass flows and concentration levels in SimpleBox is done with concentration-based “piston velocity”

type mass transfer coefficients.

The structures of the reviewed models are summarized and compared in Table 2.1. Examination of the model structures reveals that all the models are constructed on the fugacity concept with different compartments of interest, while the differences between these fugacity-based multimedia models arise mainly in the following areas:

1. The way of treating environmental media;
2. The decision to approach inter-media transfer mechanics;
3. The approach to the fugacity concept.

Consequently, although all the models use similar well-accepted expressions to describe chemical transformation and transit, they handle the modelling development differently; they can thus yield estimates of non-equilibrium concentrations in multimedia well-mixed box environments under steady-state conditions and in some cases under unsteady-state conditions, and they have diverse modelling functions.

Table 2.1 A survey of multimedia models

Model Names	CHEMFANCE	ChemRange	ChemCAN	CalTOX	BETR	SimpleBox
Developer	Devillers et al. (1995)	Swiss Federal Institute of Technology Zürich Laboratory of Chemical Engineering	Canadian Environmental Modelling Centre (CEMC)	The California Department of Toxic Substances Control	Environmental Research Group, University of Milan at Varese,	National Institute of public Health and the Environment (RIVM)
Main Application Field	Assist in understanding chemical fate and transport in France	Assist in prediction of persistence and spatial range of organic chemicals	Assist human exposure assessment, designed for use in Canada	Exposure model for hazardous waste sites	Modelling the movement of persistent organic pollutants on a continental scale	Assist in understanding chemical fate in urban area
Spatial Resolution	No	No	No	No	Pseudo spatial	No
Spatial Scale	Regional	Global	Regional	Regional	Continental	Regional, continental and global
Temporal resolution	Steady state	Steady state	Steady state	Steady state	Steady state	Steady state and Unsteady state
Environmental Compartments	Air, water, soil, sediment,	Air, water, soil, and sediment.	Air, water, soil, and sediment.	Air, water, soil, sediment, Terrestrial vegetation	Atmosphere, water, soil, sediment, vegetation	Air, water, soil, sediment, aquatic biota, suspended solids
Modelling Format	Excel spreadsheet	Excel spreadsheet	Excel spreadsheet	Excel spreadsheet	Excel spreadsheet	Excel spreadsheet



### 2.1.3 Discussion

Fugacity-based compartmental models are widely used because of their easy execution and their convenient method of describing and quantifying the chemicals in environmental media. However, they do suffer from some limitations that restrict the modelling application, limitations that are briefly explained as follows:

1) The model parameters are based on average values. In the model, all the processes of concern take place at constant and identical rates at all places. As a result, the outputs are the average concentrations in the environmental compartments.

2) The fugacity model has limitation for the prediction of concentrations from dynamic sources. On the assumption of a “well-mixed” compartment, only average concentrations of each compartment can be estimated. On the contrary, the spatial and temporal variation is a concerned issue for the pollution caused by a point source. Accordingly, the interpretation of the averaged concentration will lead to misunderstanding in environmental management (Cohen 1986).

3) Compartmental model is not appropriate for modelling the fate of chemicals quantitatively at a small scale. At the smaller scales, the key assumption of the uniform mixing of the chemicals in the environmental media becomes invalid. The model is used only to gain a qualitative understanding of the processes affecting a chemical's fate at theses smaller scales (Cohen 1986).

## **2.2 ISMCM Model**

### **2.2.1 Model Description**

Cohen and his co-workers have applied the concept of multimedia compartment modelling as a screening tool in the development the Multimedia Compartment Model (MCM) (Cohen and Ryan 1985), which has been followed by the Spatial Multimedia Compartment Model (SMCM) (Cohen et al. 1990), and more recently by the ISMCM (Cohen and Cooter 2002), which allows for non-uniformity in some compartments. Thus, only ISMCM is illustrated in this brief review.

ISMCM considers all media, biological and non-biological, in one integrated system. It combines coupled uniform and non-uniform media by using intermedia physical boundary conditions to account for the complex transport of pollutants through the ecosystem, in which soil and sediment compartments are treated as non-uniform media, while the others are taken to be uniform.

ISMCM applies the concept of fugacity capacity to uniform compartments and takes into consideration mass balance under unsteady state. On the other hand, a 1-D convective-diffusion transport process is described in the non-uniform compartments. Spatial resolution, therefore, can be obtained in the soil and sediment compartments, whereas spatial resolution is sacrificed in the uniform compartments. Assuming mass conservation, ISMCM is able to predict transport based on a sound mechanistic

description of environmental processes, including the estimation of intermedia transfer factors.

### **2.2.2 Discussion**

ISMCM is a hybrid model of combining compartmental model and single-medium model in an attempt to provide a screening-level tool. It avoids the oversimplification of compartmental model and can provide some spatial resolution that is not found in compartmental model. However, it is not comparable with LSSMM with respect to the resolutions of outputs, especially, in any case where the spatial and/or temporal resolution of prediction plays a significant role in the evaluation of risk assessment for human health, or in a simulation occurring on a small scale.

## **2.3 LSSMM Model**

### **2.3.1 Model Description**

LSSMM is composed of serially linked single-medium transport models. The concentration of a chemical in a specific medium in which a pollutant source exists is calculated first, then the inter-media transfer rates are calculated and used as source inputs to calculate the contaminant concentration in other adjoining media.

### **2.3.2 Variants of LSSMM**

The Multimedia Environmental Pollutant Assessment System (MEPAS), developed by the Pacific Northwest Laboratory, is an integrated analytical model, which is specifically designed to address general problems at any hazardous waste site (Droppo et al. 1989).

MEPAS can simulate the transport and distribution of chemical and radiological contaminants over time and space within environmental media including air, soil, groundwater, and surface water. It estimates long-term health effects at reception locations, from exposures over 70 years, and determines acute effects from normalized maximum hourly concentrations (Droppo et al. 1989).

MEPAS, which includes nine source types, has varied capability for source term modelling. The release rate from the source term is based on the empirical equation calculated, for example, from the contaminant's vapour pressure, soil/ vapour partitioning and molecular diffusion in the air, or it is provided by the user (USEPA 1996, Laniak et al. 1997). For the atmospheric component, MEPAS has a distinct option of back-calculation release rates from measured concentrations at the receptor.

In the air transport module, MEPAS uses a sector-averaged Gaussian plume equation for air transport. Volatilisation is calculated with either steady state or transient equations, depending on the scenario of release (USEPA 1996).

Contaminant concentrations in the unsaturated soil zone can be input in the case of initially contaminated soil or it can be calculated using source terms given by the user.

MEAS uses a one-dimensional, semi-analytical transport model that assumes a constant vertical velocity for each layer in the vadose zone. Therefore, the heterogeneity in the vadose zone can be modelled by using multiple vadose zone layers with varying velocity between layers and with the infiltration rate changing with time (Droppo et al. 1989, USEPA 1996).

MEPAS simulates the transport of contaminants in the saturated groundwater flow, using a three-dimensional, semi-analytical transport model. Groundwater velocity is assumed to be uniform and horizontal, and thus one-dimensional. Water concentrations at various receptor wells can be calculated in this module; consequently, they can be used in the exposure module.

The surface water module of MEPAS allows for the simulation of rivers and wetland environments.

The various modules that constitute the model are based on well-known approaches from the contaminant transport literature. These modules allow for spatial discrimination and incorporate a number of important transport pathways.

Similarly, MMSOILS (the Multimedia Contaminant Fate, Transport, and Exposure Model) has been developed by the U.S. EPA for the evaluation of hazardous waste sites (USEPA 1988, 1996). It was designed specifically to simulate the release of toxic chemicals from underground storage tanks, surface impoundments, waste piles, and landfills. Compared to some models, like MEPAS, which have the capability to simulate the fate and transport of radioactive contaminants, MMSOILS models chemical transport

and transformation only. MMSOILS has many similar scenarios and calculation algorithms to those in MEPAS, for instance, the scenarios of releases from landfill, the algorithm of volatilisation in the air transport, and a similar approach to modelling the transport of contaminants in the saturated zone. MMSOILS, however, has an advantage in the unsaturated zone. It provides a complex model, using a one-dimensional, finite-element flow and transport model that allows for layered heterogeneity in the unsaturated zone. So far as overall capability is concerned, it has less simulation capability and application variability than MEPAS.

Third, the computer program RESRAD (RESidual RADioactivity) has been developed by the U.S. Department of Energy (DOE) and Argonne National Laboratory (ANL) to evaluate the clean up and remediation of radionuclide-contaminated soils at DOE facilities. RESRAD is similar to the MEPAS and MMSOILS but differs in constructs with respect to simulating direct release to the various media. A benchmarking analysis for these three models is given by Laniak et al. (1997) and Mills et al. (1997). The detailed difference between three models can be referred to these literatures. General differences associated with the models are presented here. RESRAD simulates the formation, decay, and transport of the products of radionuclide decay, allowing the decay products to move at a different speed than their parent.

In the unsaturated and saturated zone, RESRAD is based on a non-dispersive model that transports contaminants via advection only, unlike the previous two models, which are based on the analytical solutions to the advective-dispersive equation.

Finally, the Multimedia, Multipathway, and Multireceptor Risk Assessment (3MRA) modelling system has been designed and developed by the U.S. Environmental Protection Agency (EPA) Office of Solid Waste (OSW) and the EPA's Office of Research and Development (ORD) with the intention of supporting national assessments and regulatory actions (USEPA 2003). The 3MRA modelling system simulates contaminant releases from a waste management unit (WMU) to the various media (air, water, soil) based on the physical-chemical properties of the constituent, the characteristics of the modelled WMU, and the environmental setting (e.g., meteorological region) in which the facility is located.

Five kinds of WMU, such as landfills, waste piles etc., are considered in the source term of the 3MRA multimedia modelling system. The fate and transport model simulating the source term is modified from Jury's model (Jury et al. 1983, 1990). Thus, there is no need to use the empirical equation to estimate the chemical volatilisation from the WMU surface, which is a feature that is largely different from the previous studies.

For the unsaturated zone and saturated zone modules, 3MRA uses models similar to those used by the earlier studies.

Although 3MRA removes the limitation in the approximation of gaseous emission from a WMU cover, the contaminant transfer fluxes between media are estimated as steady state, annual average fluxes. Thus, the modelling outcomes might be in pseudo temporal, spatial resolutions that are not close to the transport process in the "real world".

### 2.3.3 Discussion

LSSMM has a powerful advantage, namely a high degree of spatial resolution, to which compartmental model and ISMCM cannot be compared. This strength makes LSSMM suitable not only for application on a large scale and on the long-term screening level, but also for a small-scale and short-term prediction. In particular, for assessing the impacts caused by toxic chemicals in the vicinity of a hazardous waste site, LSSMM has unique and overwhelming advantages over the other models.

In the previous studies of LSSMM, this review indicates that it needs extensions as described on below:

- 1) Empirical algorithms are used to estimate the organic compound volatilisation from the soil surface. Therefore, the developed modules are suitable for one case but may not be appropriate for dealing other similar sites. As a result, the chemical mass balance is difficult to be maintained with this kind of modelling process. An additional mass balance testing is required; otherwise, the mass conservation cannot be guaranteed.

- 2) The pollutant transfer rates are estimated as the steady-state annual average rate. This simplified approximation obviously differs greatly from the real transport process.

- 3) The use of average concentrations and fluxes precludes the determination of dynamic effects.



## 2.4 SUMMARY

The representative models of three categories of multimedia models, namely the compartmental model, ISMCM, and LSSMM, are reviewed in this chapter. The literature review indicates that some shortcomings exist in the previous studies of multimedia models. These limitations include the following:

1. The models use an oversimplified way to treat all or some of the complex intermedia transport processes, such as volatilisation and leachate release, thereby leading to the limited modelling application only for large-scale, long-term risk assessment at the screening level (e.g. compartmental models and ISMCM);
2. Steady state methods are employed to estimate pollutant releases from contaminated site, for example, MEPAS and MMSOILS;
3. The models require a large number of input parameters, which increase the level of complexities, for instance, MEPAS and MMSOILS;
4. The complexity of model execution and the requirement that the user be able to implement the modelling system are ubiquitous challenges for the existing models, for instance, 3MRA, MMSOILS.

With the recent progress towards a better understanding of the dynamics and complexities of environmental multimedia pollution issues, and because of the increased availability of mathematical and computational tools, it is possible to extend previous

efforts on environmental multimedia modelling in the following areas:

1. Identifying the state-of-the-art mechanistic approaches to compute the inter-media fluxes;
2. Providing more realistic simulations of contaminant releases in the source term scenario;
3. Providing more realistic simulations of contaminant dispersion from the source term into a multimedia environment;
4. Modest model input requirement to accomplish the reasonable perditions;
5. Easily accessed uncertainty analysis method to be embedded in EMMS; and
6. Easily implemented user interface shelled to EMMS.

## **Chapter 3**

# **Development of an Environmental Multimedia Modelling System**

### **3.1 Development of a Conceptual Model Framework**

Once a contaminant is released into the environment from a pollution source, such as an oil spill or a solid waste site, it will migrate all connected environmental media simultaneously and finally be exposed to humans through each pathway. Figure 3.1 illustrates such contaminant transport routes from a typical contaminated site, namely a landfill site, in which three basic environmental media are contaminated: the atmosphere, the soil, and the groundwater.

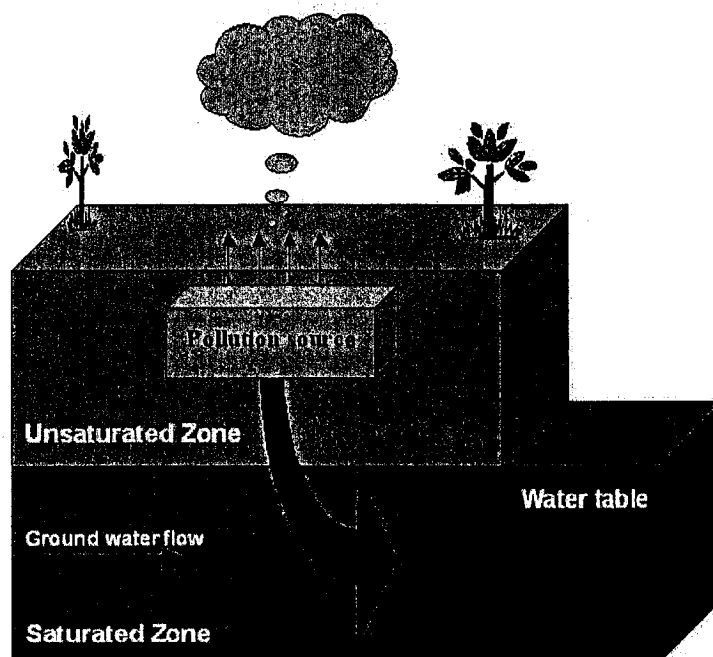


Figure 3.1 The typical multimedia environmental pollution site

To extend the previous efforts on the environmental multimedia models, an environmental multimedia modelling system (EMMS) is developed in this chapter. The conceptual framework is depicted in Figure 3.2, which shows that the modelling process begins with the release of a contaminant from a contaminated site into a multimedia environment, and ends with a concentration prediction at the risk receptor.

The developed EMMS concentrates on examining the chemical mass exchange in the environmental media, characterizing the potential risk to human health as a result of the contamination of the air and groundwater by the volatile organic compound (VOC) pollutant present at the other site. Four modules, namely the landfill module, the unsaturated zone module, the saturated zone module, and the air transport module, are

proposed in the development a new EMMS that simulates contaminant migrations from a pollution source through interconnected exposure pathways.

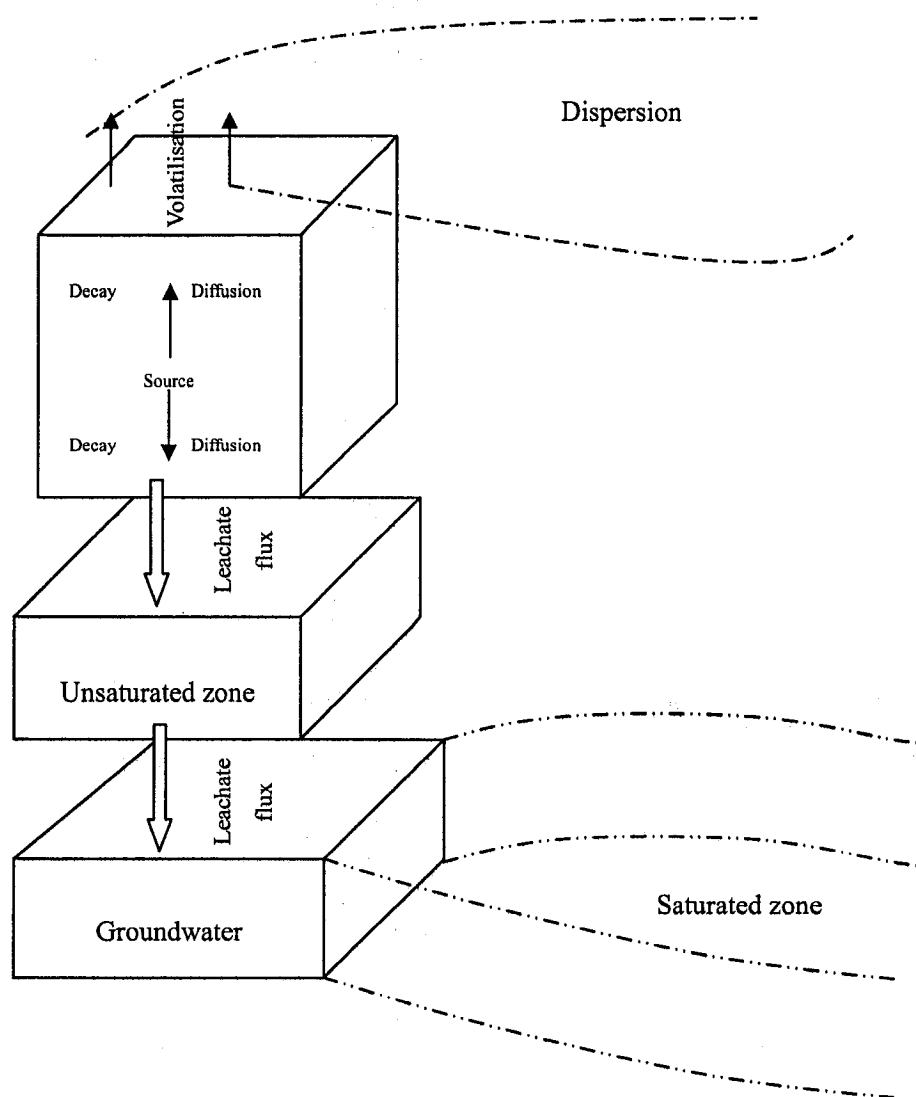


Figure 3.2 A conceptual modelling framework of the environmental multimedia modelling system

A key consideration in the development of an environmental multimedia modelling system is to provide the capacity to evaluate the risk of exposure to the contaminants released from an influential source term. Typical multimedia pollution problems usually involve organic compounds, which can evaporate, disperse, and

transport into different environments. In this study, a specific source term represented by landfill module is developed as the starting module. The concept of “landfill” in “landfill module” can be applied to generally contaminated sites not only to landfill. Along with other modules, the development of key EMMS components is explained as follows:

### 1. Landfill Module

The landfill module serves as a starting model of EMMS. It is designed to provide the assessment of contaminant mass emission rates up into the air and released leachate down into soil beneath the landfill, and to govern mass balance between the source and all affected media.

The landfill module describes the pollutant fate and transport in a porous landfill medium, in which the sorption, degradation, advection and diffusion processes are considered. This module can also be applied to other porous media, such as contaminated soil.

From the landfill module, the contaminant mass emission rate for the air and atmosphere can be computed, technically linking to an air dispersion module. For the purpose of linking the source module to the unsaturated zone module as shown in Figure 3.2, a pollution leachate generation modelling component is derived on the basis of the work of Sterile (1988) and Labieniec (1994). It moves the polluting source to the unsaturated zone module.

## 2. Unsaturated Zone Module

The unsaturated zone module simulates the movement of leachate contaminants in the subsurface after they are emitted from the base of the landfill. A three-dimensional fate and transport model is used to achieve this goal. Module outputs include the contaminant concentration in the water table and the mass flux entering into the groundwater that is used by the saturated zone module as the contaminant source.

A feature of this environmental multimedia modelling system is that the contamination source from the landfill bottom is treated as a time-dependent releasing source in the unsaturated zone module, which is a technical consideration towards a realistic simulation of landfill dynamics.

Leachate migrates vertically through the unsaturated zone and finally reaches the saturated groundwater. A leachate flux modelling component from the unsaturated zone is derived based on the concept in Carnhan and Remer (1984) and links to the saturated zone module as the contamination source. The conceptual mechanism is depicted in Figure 3.2.

## 3. Saturated Zone Module

The contaminant coming from the unsaturated zone module is presumed to be uniformly mixed in the groundwater to form a mixing zone, which serves as the source term of the saturated zone dispersion module. The saturated zone module simulates the fate and transport of dissolved contaminants from the water table to the downgradient with an

analytical solution to three-dimensional groundwater model. Module outputs are the groundwater contaminant concentrations in the potential receptors. They are used to determine whether the concentrations of concerned chemicals exceed the water standard at the downstream receptors.

#### 4. Air Dispersion Module

The air dispersion module takes the emission rate of the released pollutants, mostly VOCs, from the landfill module. It consists of two sub-models. One is the on-site concentration model; the other is the simplified Gaussian plume model. The emission rate to air from the landfill module provides the source term so that the concentration distribution in the atmosphere can be simulated. Module outputs represent the chemical concentration in the ambient atmosphere above ground.

Four modules work together under a conceptual framework to simulate the fate and transport of pollutants from a source contaminating the surrounding soil, groundwater, and air environment. The developed EMMS can be adapted to address other typical multimedia environmental pollution sites besides landfill.

### **3.2 Model Derivation and Integrated Development**

Therefore, a new EMMS is proposed and conceptually visualised in Figure 3.3. The technical consideration and derivation of contaminant fluxes between adjacent media are



the essential processes in the modelling system development. The detailed model derivations are described in the following subsections.

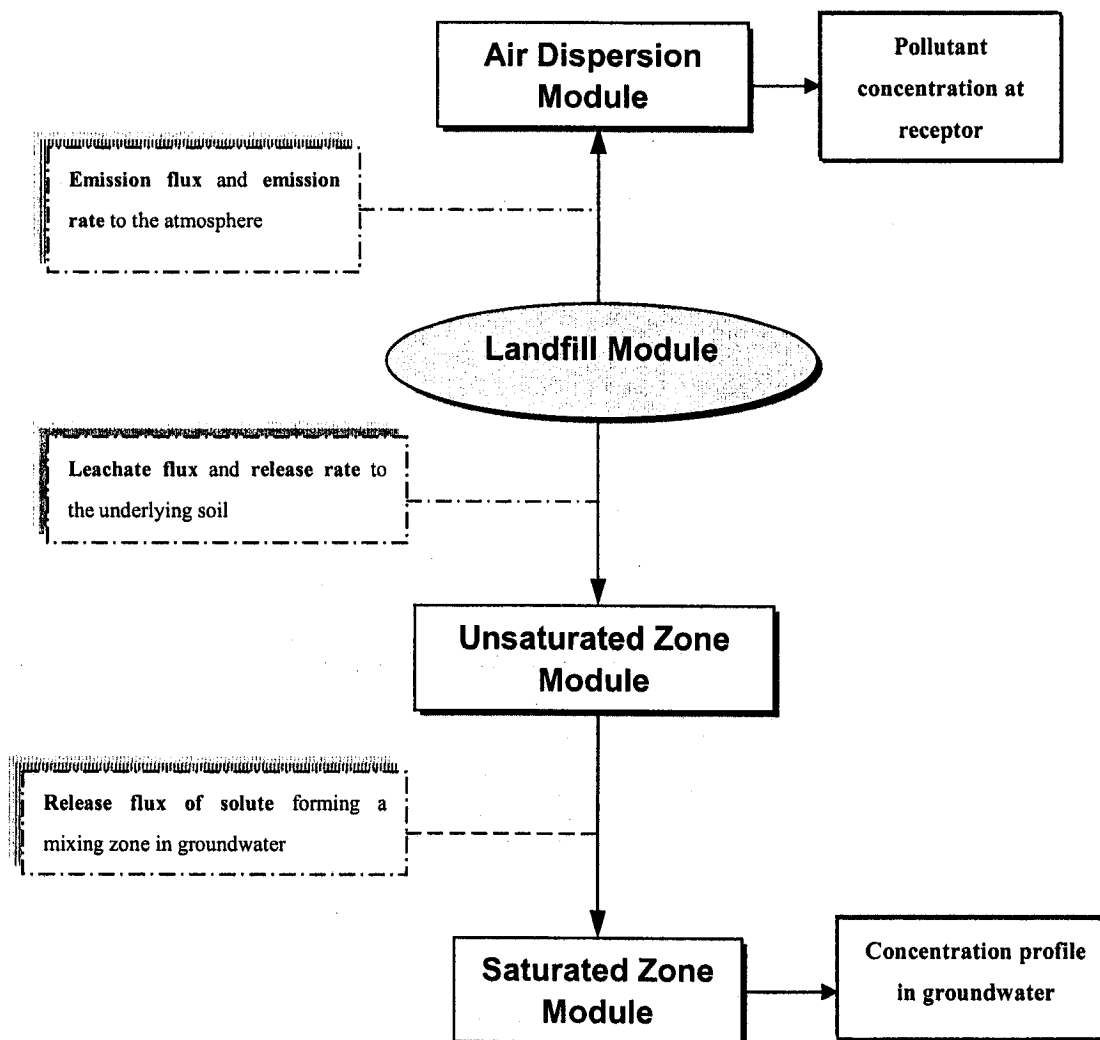


Figure 3.3 Key modules of the modelling system

### 3.2.1 Landfill Module

The landfill module is considered to simulate the dynamics of the solid waste in the

landfill chamber or the contaminated soil and to calculate the mass emission rate to air due to volatilisation as well as the leachate release rate down to the unsaturated zone. A 1-D partial differential equation is employed and solved to describe the chemical fate and transport in the landfill chamber. The solution represents the vertical concentration profile over time of the total (sorbed, dissolved, and gaseous) pollutants in the soil or waste column. From this solution, the landfill module allows an integrated consideration of the volatilisation rate of the gas-phase contaminant mass from the landfill chamber to the air and the releasing mass flux of the aqueous-phase pollutant (e.g., leachate) by advection and diffusion from the bottom of the landfill or leaking tank to the unsaturated zone.

### **3.2.1.1 Model overview and assumptions**

In the development, it is assumed that a uniform chemical concentration is initially distributed in the landfill refuse or polluted soil column. Meanwhile, the following mechanisms are considered: the transport of the chemical of interest in the vapour phase as well as in the liquid phase, the linear equilibrium partitioning among the phases, the degradation of the chemical into other compounds, and the constrained mass transfer through the top soil cover (Jury et al. 1990, Lin and Hildemann 1995).

In addition, the following assumptions have been made in the development of the landfill module (Jury et al. 1990, Lin and Hildemann 1995).

### 1. Landfill Chamber

The landfill site is conceptualized as a one-dimensional, two-chamber system: the lower compartment (of depth  $L$ ) contains contaminants, and the upper compartment of compact cover (with depth  $d$ ) separates the lower chamber from the atmosphere (Lin and Hildemann 1995). The soil in the landfill is assumed to be a homogeneous porous medium with uniform properties such as the volumetric air content  $a$ , the volumetric water content  $\theta$ , and the bulk density  $\rho_b$  ( $\text{kg/m}^3$ ). The total porosity  $\phi$  is equal to the gas-filled porosity plus the liquid-filled porosity:

$$\phi = \theta + a \quad [3.1]$$

### 2. Phases

Chemicals reside in three phases in a landfill: in an adsorbed phase, the concentration of which  $C_s$  is expressed in  $\mu\text{g/g}$  (of soil); a dissolved phase, the concentration of which  $C_L$  is expressed as  $\text{g/m}^3$  (of soil solution); and a gaseous phase, the concentration of which  $C_G$  is expressed as  $\text{g/m}^3$  (of air). The total concentration  $C_T$  in  $\text{g/m}^3$  is thus

$$C_T = \rho_b C_s + \theta C_L + a C_G \quad [3.2]$$

The movement of a contaminant, other than molecular-scale gaseous and aqueous diffusion, is modelled as a one-dimensional countercurrent flow: the gas is moving upward with an interstitial velocity  $v'_G$ , whereas the leachate is moving downward with the velocity  $v'_L$ . Sorbed phase contaminants are considered to be immobile.

### 3. Partitioning

The contaminant undergoes reversible, linear equilibrium partitioning between the adsorbed and the dissolved phases, and between the adsorbed and the gaseous phase.

Soil-liquid: 
$$C_S = K_D C_L \quad [3.3]$$

where  $K_D$  is the liquid-soil adsorption coefficient ( $\text{m}^3/\text{kg}$ ).

For organic constituents, 
$$K_D = f_{oc} K_{oc} \quad [3.4]$$

where  $f_{oc}$  is the organic carbon fraction in the soil or waste (dimensionless);  $K_{oc}$  is the equilibrium partition coefficient, normalized to organic carbon ( $\text{m}^3/\text{kg}$ ).

soil-gaseous: 
$$C_S = K_G C_G \quad [3.5]$$

where  $K_G$  is the gas-soil adsorption coefficient ( $\text{m}^3/\text{kg}$ ).

It is implicit in this linear equilibrium partitioning assumption that the sorptive capacity of the soil column solids is considered to be infinite with respect to the total mass of the constituent over the duration of the simulation, i.e., the soil column sorptive capacity does not become exhausted.

Contaminants in the dissolved and gaseous phases are assumed to be in equilibrium and to follow Henry's law.

$$C_G = K_H C_L \quad [3.6]$$

where  $K_H$  is dimensionless Henry's law constant.

#### 4. Fluxes

The Chemical flux  $J_i$  ( $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ) is the sum of the vapour flux and the flux of dissolved solute.

## 5. Others

1). The contaminant mass may be lost from the soil column through one or more first-order loss processes.

2). The effective diffusivity in the soil may be calculated according to the model of Millington and Quirk (1961)

$$D_g = \frac{\theta^{10/3} D_g^a}{\phi^2} \quad [3.7]$$

where  $D_g$  and  $D_g^a$  are the gaseous diffusion coefficients in the soil and air, respectively ( $\text{m}^2/\text{d}$ ).

$$D_l = \frac{\theta^{10/3} D_l^w}{\phi^2} \quad [3.8]$$

where  $D_l$  and  $D_l^w$  are the liquid diffusion coefficients in the soil and pure water, respectively ( $\text{m}^2/\text{d}$ ).

Under these assumptions above, the individual concentration in each phase ( $C_G, C_L, C_S$ ) can be expressed as a function of the total concentration  $C_T$ , which has been chosen as the only dependent variable.

$$C_T = R_S C_S = R_G C_G = R_L C_L \quad [3.9]$$

where

$$R_S = \rho_b + \frac{\theta}{K_D} + \frac{aK_H}{K_D} \quad [3.10]$$

$$R_L = \rho_b K_D + \theta + aK_H \quad [3.11]$$

$$R_G = \frac{\rho_b K_D}{K_H} + \frac{\theta}{K_H} + a \quad [3.12]$$

$R_s$   $R_L$   $R_G$  are the equilibrium distribution coefficient between the total and solid, aqueous, and gaseous phase contaminant concentrations, respectively.

### 3.2.1.2 Governing equation and solution

Then, the overall governing equation for the pollutant fate and transport become:

$$\frac{\partial C_T}{\partial t} = D_E \left( \frac{\partial^2 C}{\partial z^2} \right) - V_E \left( \frac{\partial C}{\partial z} \right) - \mu C_T \quad [3.13]$$

where  $\mu$  is the effective first order reaction rate constant ( $d^{-1}$ ) and can be calculated by

$$\mu = \frac{\ln(2)}{\tau} \quad [3.14]$$

in which  $\tau$  is defined as the effective half-life in the unsaturated zone (d).

$V_E$  is the effective solute velocity (m/d) and is equal to

$$V_E = -\frac{av'_G}{R_G} + \frac{\theta v'_L}{R_L} = -\frac{v_G}{R_G} + \frac{v_L}{R_L} \quad [3.15]$$

in which  $v_G$  and  $v_L$  are the bulk gas and water velocities (m/d), respectively.

$D_E$  is named the effective diffusion coefficient ( $m^2/d$ ); it is transformed from

$$D_E = \frac{a^{10/3} D_g^a K_H + \theta^{10/3} D_l^w}{\phi^2 R_l} \quad [3.16]$$

Under the initial condition

$$C_T(z,0) = C_{L0} \quad \text{if } 0 < z < L \quad [3.17]$$

$$C_T(z,0) = 0 \quad \text{if } z > L \quad [3.18]$$

in which  $C_{L0}$  is the background concentration in landfill; and upper boundary condition

$$J_i(0,t) = -hC_G(0,t) \quad [3.19]$$

where  $h$  is the overall mass transfer coefficient through the top cover (m/d) and is estimated by:

$$\frac{1}{h} = \frac{d}{D_g} + \frac{1}{k_i} \quad [3.20]$$

in which  $k_i$  is the mass transfer coefficient in the air-soil boundary layer (m/d).

Therefore, the analytical solution can be obtained in the equation below (Lin and Hildemann 1995):

$$\begin{aligned} C_T(z,t) = & \frac{1}{2} C_{L0} \exp(-\mu t) \left\{ \operatorname{erfc} \left[ \frac{z-L-V_E t}{\sqrt{4D_E t}} \right] - \operatorname{erfc} \left[ \frac{z-V_E t}{\sqrt{4D_E t}} \right] + \right. \\ & \left( 1 + \frac{V_E}{H_E} \right) \exp \left( \frac{V_E z}{D_E} \right) \left[ \operatorname{erfc} \left( \frac{z+L+V_E t}{\sqrt{4D_E t}} \right) - \operatorname{erfc} \left( \frac{z+V_E t}{\sqrt{4D_E t}} \right) \right] + \\ & \left( 2 + \frac{V_E}{H_E} \right) \exp \left[ \frac{H_E(H_E+V_E)t + (H_E+V_E)z}{D_E} \right] \\ & \left. \left[ \operatorname{erfc} \left( \frac{z+(2H_E+V_E)t}{\sqrt{4D_E t}} \right) - \exp \left( \frac{H_E L}{D_E} \right) \operatorname{erfc} \left( \frac{z+L+(2H_E+V_E)t}{\sqrt{4D_E t}} \right) \right] \right\} \end{aligned} \quad [3.21]$$

$$\text{where } H_E = \frac{hK_H}{R_i} \text{ with unit (m/d)} \quad [3.22]$$

### 3.2.1.3 Emission rate generation and its linkage to air dispersion module

The contaminant volatilises to the air from the landfill surface. The emission flux of pollutant can be computed by (Lin and Hildemann 1995):

$$J_i(0,t) = \frac{C_{L0} \exp(-\mu t)}{2} \left\{ V_E \left[ \operatorname{erfc} \left( \frac{V_E t}{\sqrt{4D_E t}} \right) - \operatorname{erfc} \left( \frac{L + V_E t}{\sqrt{4D_E t}} \right) \right] + \right. \\ \left. (2H_E + V_E) \exp \left[ \frac{H_E (H_E + V_E) t}{D_E} \right] \left[ \exp \left( \frac{H_E L}{D_E} \right) \operatorname{erfc} \left( \frac{L + (2H_E + V_E) t}{\sqrt{4D_E t}} \right) - \operatorname{erfc} \left( \frac{(2H_E + V_E) t}{\sqrt{4D_E t}} \right) \right] \right\} \quad [3.23]$$

The cumulative mass lost from the landfill surface per unit area of contamination site due to volatilisation,  $M_{vol}(\text{g/m}^2)$ , is (Jury et al. 1990).

$$M_{vol}(t) = \int_0^t -J_i(0,t') dt' \quad [3.24]$$

The time-weighted average of the mass emission rate from the surface of the site to the atmosphere over the annual duration,  $Q(t_{ad})(\text{g/s})$ , is used in the air dispersion module and is calculated by:

$$Q(t_{ad}) = \frac{M_{vol}(t_{ad}) A_x A_y}{t_{ad} 86400} \quad [3.25]$$

where  $t_{ad}$  is the annual exposure duration (d);  $A_x$  and  $A_y$  are the length and width of the landfill area (m), and x-axis is the direction of groundwater flow.

#### 3.2.1.4 Leachate generation and its linkage to unsaturated zone module

The mass transport of an organic pollutant in the landfill chamber can be expressed as the flux in the gas phase and the aqueous phase (Jury et al. 1990; Labieniec 1994).

The gas phase diffusive flux ( $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ) is calculated by:



$$J_g(z,t) = \frac{-a^{10/3} D_g^a K_H}{\phi^2 R_L} \cdot \frac{\partial C_T(z,t)}{\partial z} \quad [3.26]$$

The aqueous phase diffusive flux ( $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ) is computed by:

$$J_l(z,t) = \frac{-\theta^{10/3} D_l^w}{\phi^2 R_l} \cdot \frac{\partial C_T(z,t)}{\partial z} \quad [3.27]$$

And the aqueous and gaseous phase advective flux ( $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ) is equal to

$$J_a(z,t) = V_E C_T(z,t) \quad [3.28]$$

The total mass flux ( $\text{g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ) is the sum of the three equations above, calculated by:

$$J_t(z,t) = J_g(z,t) + J_l(z,t) + J_a(z,t) \quad [3.29]$$

Thus, the cumulative mass per unit area of the contaminated site that has moved across a horizontal plane at depth  $z$ , and time,  $t$ , due to each of the mass transport processes is determined by the following equations (Labieniec 1994).

The cumulative gas phase diffusive flux ( $\text{g}/\text{m}^2$ ) is predicted by using:

$$M_g(z,t) = \int_0^t J_g(z,t') dt' \quad [3.30]$$

The cumulative aqueous phase diffusive flux ( $\text{g}/\text{m}^2$ ) is estimated with:

$$M_l(z,t) = \int_0^t J_l(z,t') dt' \quad [3.31]$$

And the cumulative aqueous phase advective flux ( $\text{g}/\text{m}^2$ ) is determined by

$$M_a(z,t) = \int_0^t J_a(z,t') dt' \quad [3.32]$$

Finally, the total cumulative flux ( $\text{g}/\text{m}^2$ ) is calculated by

$$M_t(z, t) = M_g(z, t) + M_l(z, t) + M_a(z, t) \quad [3.33]$$

### 3.2.2 Unsaturated Zone Module

The unsaturated zone module simulates the vertical transport of contaminants in the leachate from the base of the landfill to the lower unsaturated zone boundary at the water table, and estimates the contaminant flux to the saturated zone module. For this transport pathway, most multimedia models simplify the unsaturated zone to a one-dimensional model that assumes that the contaminant migrates only vertically from the waste source to the underlying water table. In most cases, this is a valid assumption because the scale of transport in the unsaturated zone tends to be orders of magnitude smaller than in the saturated ground water (USEPA 1996). However, the contribution of lateral diffusion to this system cannot be neglected. Therefore, a 3-D transport model is proposed to address the unsaturated zone dynamics.

#### 3.2.2.1 Key assumptions

The following assumptions are used in constructing the unsaturated zone module.

- Contaminants are released from a square waste site; there is no contaminant flux outside the waste site.
- Flow is mainly vertical.

- Flow and transport are steady-state
- The soil is initially free of contamination.
- Soil is a uniform porous medium with uniform properties;
- Contaminant transport is by advection and dispersion.
- Contaminants are in the aqueous and sorbed phases; there are no mass transfer processes between phases other than adsorption onto soil particles.
- There is no volatilisation from the unsaturated zone (i.e., no gas-phase release or transport).

### 3.2.2.2 Governing equation

A 3-D solute transport analytical solution is proposed for the unsaturated zone (Carnhan and Remer 1984):

$$\frac{\partial C_{un}}{\partial t} = \frac{D_T}{B_1} \left( \frac{\partial^2 C_{un}}{\partial x^2} + \frac{\partial^2 C_{un}}{\partial y^2} \right) + \frac{D_L}{B_1} \frac{\partial^2 C_{un}}{\partial z^2} - \frac{v}{B_1} \frac{\partial C_{un}}{\partial z} - \mu_{unsat} C_{un} \quad [3.34]$$

$$Q = K_D C_{un} \quad [3.35]$$

$$B_1 = 1 + \alpha K_D \quad [3.36]$$

$$\alpha = (1 - \phi_{un}) / \phi_{un} \quad [3.37]$$

where  $C_{un}$  is the concentration of solute in the fluid phase ( $\text{g/m}^3$ );  $Q$  is the concentration of the solute in the solid phase ( $\text{g/m}^3$ );  $K_{Dl}$  is the liquid-soil adsorption coefficient ( $\text{m}^3/\text{kg}$ );  $D_L$  is the coefficient of longitudinal dispersion ( $\text{m}^2/\text{d}$ );  $D_T$  is the coefficient of

transverse dispersion ( $\text{m}^2/\text{d}$ );  $v$  is the average velocity of the fluid ( $\text{m}/\text{d}$ );  $\phi_{un}$  is the effective porosity in the unsaturated zone;  $\alpha$  is the ratio of the solid volume to the fluid volume; other symbols have same meanings as previous;  $B_1$  is a dimensionless retardation factor;  $\mu_{unsat}$  is the first order reaction rate constant in the unsaturated soil ( $\text{d}^{-1}$ ), and equal to  $\frac{\ln(2)}{\tau_{unsat}}$ , in which  $\tau_{unsat}$  is the half-life in the unsaturated zone ( $\text{d}$ ).

Most multimedia models consider the nonsteady- state leachate flux from waste sites as a steady-state annual average flux. This consideration is appropriate for long-term screening but is not an accurate assessment, as it cannot reflect the peak contaminant concentration in the soil. In order to mitigate this limitation, we calculate the annual leachate flux from the landfill site, and then transform it into a continuous time-dependent release source that is the contaminating source to the unsaturated zone module.

If  $M(t)$  is the total mass of the contaminant moving out of the landfill and into the soil at time  $t$ , it is calculated as:

$$M(t) = \frac{m_0}{\gamma} [\exp(-\mu_{unsat}t) - \exp(-\mu_{unsat} - \gamma)t] \quad [3.38]$$

where  $m_0$  is rate of release of the solute from the point source at time zero ( $\text{g}/\text{d}$ );  $\gamma$  is the time constant for the decay of the release rate ( $\text{d}^{-1}$ ). It means that the leachate release from the landfill site to the unsaturated zone at  $t=0$ , and the release rate is decreasing with the decay factor  $\gamma$ . During an annual time-weighted evaluation period, the pollutant mass entering into the soil can be calculated by Equation [3.33]:  $M(365) = M_i(L, 365) A_x A_y$ ,

where  $A_x$  and  $A_y$  are the same definition as given in Equation [3.25].

Simplifying the calculation process, given  $\gamma = \frac{v^2}{4D_L B_1}$ ,  $m_0$  can be back calculated based on Equation [3.38].

Provided that the net change of pollutant mass  $M(t)$  is (Carnahan and Remer 1984)

$$\frac{dM(t)}{dt} = m_0 \exp[-(\gamma + \mu_{unsat})t] - \mu_{unsat} M(t) \quad [3.39]$$

An analytical solution to Equation [3.34] can be obtained:

$$C_{un} = \frac{m_0 \exp[-(\gamma + \mu_{unsat})t]}{4\pi\phi_{un}\eta D_T D_L^{1/2}} \exp\left(\frac{vz}{2D_L}\right) \operatorname{erfc}\left[0.5\eta\left(\frac{B_1}{t}\right)^{1/2}\right] \quad [3.40]$$

$$\text{where } \eta = \left(\frac{r^2}{D_T} + \frac{z^2}{D_L}\right)^{1/2}, \quad r^2 = x^2 + y^2 \quad [3.41]$$

### 3.2.2.3 Link to saturated zone model

A water table is assumed to exist below the contaminated zone at  $z=z_{wt}(m)$ . The leachate fluxes are computed a by

$$J_{un}(z,t) = \frac{-D_L}{B_1} \cdot \frac{\partial C_{un}(z,t)}{\partial z} + \frac{v}{B_1} C_{un}(z,t) \quad [3.42]$$

in which  $\partial C / \partial z$  is derived from Equation [3.40] and the solution is:

$$\begin{aligned} \frac{\partial C_{un}}{\partial z} = & \frac{m_0 \exp[-(\gamma + \mu_{unsat})t]}{4\pi\phi_{un}D_T D_L^{1/2}} \left[ -\frac{z}{D_L \eta^3} \exp\left(\frac{vz}{2D_L}\right) \operatorname{erfc}\left[0.5\eta\left(\frac{B_1}{t}\right)^{1/2}\right] \right. \\ & \left. + \frac{v}{2D_L \eta} \exp\left(\frac{vz}{2D_L}\right) \operatorname{erfc}\left[0.5\eta\left(\frac{B_1}{t}\right)^{1/2}\right] - \frac{z}{D_L \eta^2 \sqrt{\pi}} \left(\frac{B_1}{t}\right)^{1/2} \exp\left(\frac{vz}{2D_L} - \frac{B_1 \eta^2}{4t}\right) \right] \end{aligned} \quad [3.43]$$

The cumulative mass concentration crossing the horizontal plane at  $z$  depth and at time  $t$  and entering saturated zone is expressed by

$$M_{un} = \int_0^t J_{un}(z, t') dt' \quad [3.44]$$

### 3.2.3 Saturated Zone Module

#### 3.2.3.1 Aquifer mixing zone

A contaminant plume formed beneath a waste site due to the leachate percolating through the waste site and the unsaturated zone is modelled by the unsaturated zone model and will serve as the source term of the saturated zone model. It is assumed that mass entering the saturated zone from the unsaturated soil is mixed in a mixing zone, with a volume defined by  $A_x \cdot A_y \cdot H$ . The contaminant flux coming out of the mixing zone is carried by advection through the flow area  $A_y \cdot H$ . Near-source dispersion is assumed to be insignificant.  $H$ , the aquifer mixing zone thickness, is the vertical thickness of this plume within the aquifer at the point where the plume passes beneath the contamination

site (Solhotra et al 1995). It is calculated by

$$H = \sqrt{2\alpha_z A_x} + B \left( 1 - \exp \left( - \frac{A_x Q_f}{V_d \phi_s B} \right) \right) \quad [3.45]$$

where  $H$  is the aquifer mixing zone thickness (m);  $\alpha_z$  is the vertical dispersivity (m);  $A_x$  is the length of facility measured parallel to the flow direction (m);  $A_y$  is the length of orthogonal to the groundwater flow;  $B$  is the thickness of the saturated zone (m);  $V_d$  is the uniform seepage velocity in the flow (x) direction (m/yr);  $Q_f$  is the percolation rate (m/yr);  $\phi_s$  is the effective porosity of the saturated zone.

The time-weighted average contaminant concentration in the mixing zone,  $C_0$  ( $\text{g/m}^3$ ), during each of the time periods is calculated by applying a steady-state mass balance to the mixing zone with a consideration of the average contaminant flux from the unsaturated soil to the saturated zone during each of the time period.

$$C_{s0} = \frac{M_{\text{un}}(z_{\text{wt}}, t)}{t} \cdot \frac{A_x}{V_d \frac{H}{R} + \mu_{\text{sat}} H A_x} \quad [3.46]$$

$$\text{where } R = 1 + \frac{\rho_{\text{sat}} f_{\text{ocsat}} K_{\text{oc}}}{\phi_s} \quad [3.47]$$

$R$  is retardation factor (dimensionless);  $V_d$  is Darcy velocity (m/d);  $\rho_{\text{sat}}$  is the bulk density of the saturated zone ( $\text{kg/m}^3$ );  $f_{\text{ocsat}}$  is the organic carbon fraction in the saturated zone;  $\mu_{\text{sat}}$  is the first-order degradation rate ( $\text{d}^{-1}$ ) calculated by  $\ln(2)/\tau_{\text{sat}}$ , in which  $\tau_{\text{sat}}$  is the half-life in the saturated zone (d).

### 3.2.3.2 Saturated zone model

The saturated zone model builds on the solution to the three-dimensional advection-dispersion-reaction equation with a plane source at constant concentration (Domenico 1987).

$$C_s(x, y, t) = \frac{C_{s0}}{8} \exp\left(\frac{x}{2\alpha_x} \left[1 - \left(1 + \frac{4\mu_{sat}\alpha_x}{V_d R}\right)^{1/2}\right]\right) \operatorname{erfc}\left(\frac{x - (V_d t / R)(1 + 4\mu_{sat}\alpha_x R / V_d)^{1/2}}{2(\alpha_x V_d t / R)^{1/2}}\right) \\ \left\{ \operatorname{erf}\left[\frac{y + A_y / 2}{2(\alpha_y x)^{1/2}}\right] - \operatorname{erf}\left[\frac{y - A_y / 2}{2(\alpha_y x)^{1/2}}\right] \right\} \left\{ \operatorname{erf}\left[\frac{z + H}{2(\alpha_z x)^{1/2}}\right] - \operatorname{erf}\left[\frac{z - H}{2(\alpha_z x)^{1/2}}\right] \right\} \quad [3.48]$$

where,  $V_d$  is the seepage velocity (m/d), and  $\alpha_x$ ,  $\alpha_y$ ,  $\alpha_z$  are the dispersivities in the coordinate directions (m). The dispersivity in a porous medium is defined as the dispersion coefficient divided by the mean seepage velocity, where

$$\alpha_x = \frac{D_x}{V_d}, \quad \alpha_y = \frac{D_y}{V_d}, \quad \alpha_z = \frac{D_z}{V_d} \quad [3.49]$$

and  $D_x, D_y, D_z$  are the dispersion coefficient ( $\text{m}^2/\text{d}$ ) in x, y, and z directions, respectively.

The groundwater zone is assumed to be homogenous, isotropic, unconfined, and shallow with constant thickness  $H$  (m). The flow is assumed to be in a steady state with Darcy velocity,  $V_d$  (m/d), aligned with the x-axis in the positive x-direction, and orthogonal to the vertical plane passing through the down gradient edge of the site. It states that the contaminant mass degradation in the saturated zone is first-order, and sorption is at equilibrium and proportional to the dissolved concentration. The mass loss



from the saturated zone due to volatilization at the water table is not considered.

Since the solute-transport equation is a linear partial-differential equation, the principle of superposition can be used to calculate concentrations in the system if the solute concentrations at the inflow boundary vary over time. To consider the time-dependent factors associated with a changing emission source into the groundwater, the general form of the solution can be expressed as (Wexler 1992)

$$C_s = C_{s0} \cdot A(x, y, t) + (C_{s1} - C_{s0}) \cdot A(x, y, (t - t_1)) \quad [3.50]$$

where  $C_{s0}$  is the initial solute concentration at the boundary;  $A(x, y, t)$  is the general form of the analytical solution where concentration is a function of space and time;  $C_{s1}$  is the solute concentration at the boundary after  $t=t_1$ , and  $t_1$  is the time at which the solute concentration changes at the boundary.

### **3.2.4 Air Pollution Dispersion Module**

The air-dispersion pathway is one of the principal pathways through which VOCs and toxic chemicals volatilising from waste sites may reach living organisms. Subsequently, these chemicals disperse downwind and deposit on ground surfaces in a pattern dependent on the local meteorology, the location of the point of release, the nature of the terrain downwind of the release, and the physical and chemical characteristics of the emission (USEPA 1996)

The air dispersion module estimates the annual average air concentration of

dispersed constituents at various receptor points in the area of interest. This module simulates the transport and diffusion of constituents in the form of volatilized gases emitted from area sources into the air.

For the on-site contaminant concentration prediction, it is assumed that the air above the landfill site is contained in a box that has two open walls perpendicular to the wind direction (USEPA 1986; Labieniec 1994). The base is the contaminated area where the length is  $A_y$  (m), and the annual average surface wind speed is  $u$  (m/s). The time-weighted average on-site concentration in ambient air is calculated as:

$$C_{air}(t) = \frac{Q(t_{ad})}{2uA_y} \quad [3.51]$$

To simulate the dispersion of the contaminant as it travels off-site, a simplified form of the Gaussian plume equation for a ground level source with complete reflection is employed (Turner 1970):

$$C_{air}(x,t) = \sum_{\substack{16 \text{ wind} \\ \text{directions}}} \sum_{\substack{5 \text{ wind} \\ \text{speeds}}} \sum_{\substack{6 \text{ stability} \\ \text{categories}}} \frac{2.03Q(t_{ad})f_c(\phi)}{xu\sigma_z} \quad [3.52]$$

where  $f_c(\phi)$  is the frequency or the probability of occurrence of a specific wind speed, wind direction and stability category combination;  $\sigma_z$  is the vertical dispersion coefficient (m);  $u$  is the annual average surface wind speed (m/s).

It is usually difficult to obtain detailed meteorological information about  $f_c(\phi)$ , thus according to Turner's (1970) advice, a neutral wind stability class of D and a single wind speed class with average wind speed equal to  $u$  (m/s) are assumed. After this modification, Equation [3.52] becomes:

$$C_{air}(x,t) = \frac{2.03Q(t_{ad})f(\phi)}{xu\sigma_{zD}} \quad [3.53]$$

where  $f(\phi)$  is the frequency of the wind in the direction of the exposure location;  $\sigma_{zD}$  is the vertical dispersion coefficient for D stability class approximated by  $(0.0304x+1.55)$  for less than 1,000 m (Turner 1970).

In summary, a new EMMS has been proposed in the present study with a set of modules to address the dynamics of multimedia environmental pollution issues. Many considerations related to the model formulation and environmental conditions imply that a large number of uncertainties are present. Accordingly, the modelling system will be enhanced by using two methodologies: one is based on the analysis of uncertainties; the other methodology is based on fuzzy sets. The relevant theory in uncertainty analysis is given in the next section.

### 3.3 Fuzzy-Set Based Environmental Multimedia Modelling

EMMS involves a number of parameters that need to be estimated or measured on site. Even if some parameters are measured on-site, such as the hydraulic gradient and hydraulic conductivity; they are not able to fully represent the characteristics of the contamination site, for example, due to the heterogeneity of soil and the varying of the groundwater flow. Also, the properties of chemicals of interest are obtained from experiment under certain conditions. It is difficult to take that they still keep the same

values when they occur in the contamination site. Especially, there are multimedia characteristics and complexities in most contamination sites. Therefore, it is particularly important to add to the developed EMMS an effective function of quantifying uncertainties.

Uncertainty and imprecision can be approached in different ways. Traditionally, they are handled by using probability theory and statistics. However, if the vagueness and imprecision are associated with model defining, called fuzziness, they cannot be related to probability theory but can be examined by using fuzzy set or fuzzy membership function (Evans et al. 1989, Zimmermann 2001).

In addition to being suitable for dealing with the uncertainties in the estimates of input parameters, fuzzy set methodologies can permit a model builder to capture and account for the inherent vagueness in the process of model building.

Therefore, fuzzy set and fuzzy number theory can be effective in coping with the uncertainties associated with EMMS and the related multimedia environment.

### **3.3.1 A Brief Description of the Related Fuzzy Set Theory**

Before fuzzy set methodology is applied to EMMS, the relevant fuzzy set theory is briefly illustrated in the following subsection.

### 3.3.1.1 Fuzzy set and fuzzy membership function

If  $X$  is a collection of objects denoted generically by  $x$ , then a fuzzy set  $A$  in  $X$  is a set of ordered pairs:  $A = \{(x, \mu_A(x)), x \in X\}$ ,  $\mu_A(x)$  is called the membership function or grade of membership of  $x$  in  $A$  that maps  $X$  to the membership space  $[0,1]$  (Zadeh 1973, Zimmermann 2001).

A fuzzy number is defined as a fuzzy subset of a set of real numbers,  $R$ , and is convex and normal (Dubois and Prade 1980). Thus, a fuzzy number can be considered with a generalization of the interval of confidence, which is a closed interval of  $R$  that an uncertain value belongs to (Kaufmann and Gupta 1985). It is expressed as:  $A = \{x | \mu_A(x), x \in R\}$ , where  $A$  is a fuzzy number or a fuzzy subset;  $x$  is an uncertain value.

Taking into account practical application in this study, L-R fuzzy number, triangular fuzzy number, and membership function are introduced here. Dubois and Prade (1980) defined the L-R fuzzy number as:

$$\mu_A(x) = \begin{cases} L((m-x)/a) & \text{for } x \leq m, a > 0, \\ R((x-m)/b) & \text{for } x \geq m, b > 0, \end{cases} \quad [3.54]$$

$L$  is for left and  $R$  for right reference,  $m$  is the mean value of  $A$ ,  $a$  and  $b$  are called left and right spreads, respectively. Figure 3.4 describes this kind of fuzzy number.

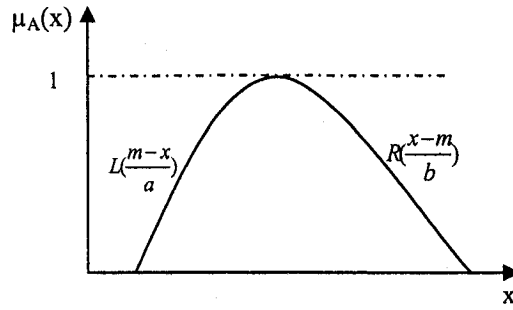


Figure 3.4 L-R fuzzy membership function

The particular case of semisymmetric L-R fuzzy number is named as triangular fuzzy number (TFN). It can be described as (Kaufmann and Gupta, 1985):

$$\mu_A(x) = \begin{cases} 0, & \text{if } x < \underline{a} \text{ and } x > \bar{a} \\ \frac{x - \underline{a}}{m - \underline{a}}, & \text{if } \underline{a} \leq x \leq m \\ \frac{\bar{a} - x}{\bar{a} - m}, & \text{if } m \leq x \leq \bar{a} \end{cases} \quad [3.55]$$

where  $[\underline{a}, \bar{a}]$  is the fuzzy interval imposed by fuzzy subset A. Also, it is depicted in Figure 3.5.

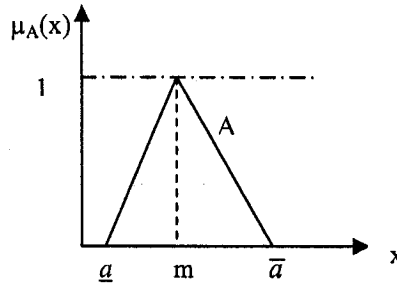


Figure 3.5 Triangular membership function

TFN is used in this study to deal with the vagueness and impression existing in the input parameters of EMMS.

To reflect the ambiguity of estimated values of a number of parameters, some important parameters that contribute to significant impacts on the modelling outcomes can be expressed in terms of fuzzy numbers. For example, the background concentration of a pollutant in a landfill described with linguistic expression ‘approximate 1.0 µg/g’ can be quantified by a fuzzy number A with the membership function  $\mu_A(x)$  when we can get a range of information that the concentration is within 0.8-1.2 µg/g.

$$\mu_A(x) = \begin{cases} 0, & \text{if } x < 0.8 \text{ and } x > 1.2 \\ \frac{x-0.8}{1.0-0.8}, & \text{if } 0.8 \leq x \leq 1.0 \\ \frac{1.2-x}{1.2-1.0}, & \text{if } 1.0 \leq x \leq 1.2 \end{cases} \quad [3.56]$$

Equation [3.56] indicates that ‘1.0 µg/g’ is the most likely value for the event that is represented by the background concentration in a site. The membership value of a fuzzy number A,  $\mu_A(x)$ , shows the possibility degree of the event that the background concentration in landfill is x. In this sense,  $\mu_A$  can be considered as a possibility distribution of the background concentration in landfill, which, in turn, can be regarded as a possibility variable restricted by the possibility distribution  $\mu_A$  (Inuiguchi and Ramik 2000).

### 3.3.1.2 Traditional operations of fuzzy number

Model input parameters will be treated as fuzzy numbers; also, the arithmetic operations

of fuzzy numbers are considered to handle the interaction of fuzzy numbers in the formulation of fuzzy-set enhanced modelling. In this context, several operations of fuzzy numbers are introduced below.

Let  $M$  and  $N$  be triangular fuzzy numbers that can be expressed as  $M = (m_M, \alpha_M, \delta_M)$  and  $N = (m_N, \alpha_N, \delta_N)$ , with  $m, \alpha, \delta$ , being the mean value, the left spread and right spread, respectively.

Operation 1-Addition of fuzzy numbers:

$$M \oplus N = (m_M + m_N, \alpha_M + \alpha_N, \delta_M + \delta_N) \quad [3.57]$$

Operation 2- Multiplication of fuzzy numbers:

Two methods are used to calculate the multiplication of two fuzzy numbers. One method approximates the multiplication of two L-R fuzzy numbers as (Dubois and Prade 1980):

$$M(\cdot)N \sim (m_M m_N, m_M \alpha_N + m_N \alpha_M, m_M \delta_N + m_N \delta_M) \quad [3.58]$$

The other is to employ the arithmetic of fuzzy numbers. Let  $\mu_M, \mu_N$  be the membership functions of fuzzy numbers  $M, N$ , respectively, multiplication can be given by (Kaufmann and Gupta, 1985):

$$\forall x, y, z \in R^+ : \mu_{M(\cdot)N}(z) = \bigvee_{z=x \cdot y} (\mu_M(x) \wedge \mu_N(y)) \quad [3.59]$$

The first method is employed in this study.

Operation 3-Multiplication between a fuzzy number and an ordinary number:

$$\begin{aligned} k \cdot M &= (km_M, k\alpha_M, k\delta_M) \quad k > 0 \\ &= (km_M, -k\alpha_M, -k\delta_M) \quad k < 0 \end{aligned} \quad [3.60]$$

in which  $k$  represents an ordinary number.



These operations on fuzzy numbers and the related theory are integrated to the modules of EMMS to quantify the related uncertainties. In this chapter, only the relevant fuzzy set theory is introduced. Further details are explained in Chapters 4, 5, and 6 through a series of case studies.

### 3.3.2 Fuzzy Set Based Environmental Multimedia Modelling

The demonstrated fuzzy set theory is incorporated into modules of EMMS as presented in Figure 3.6.

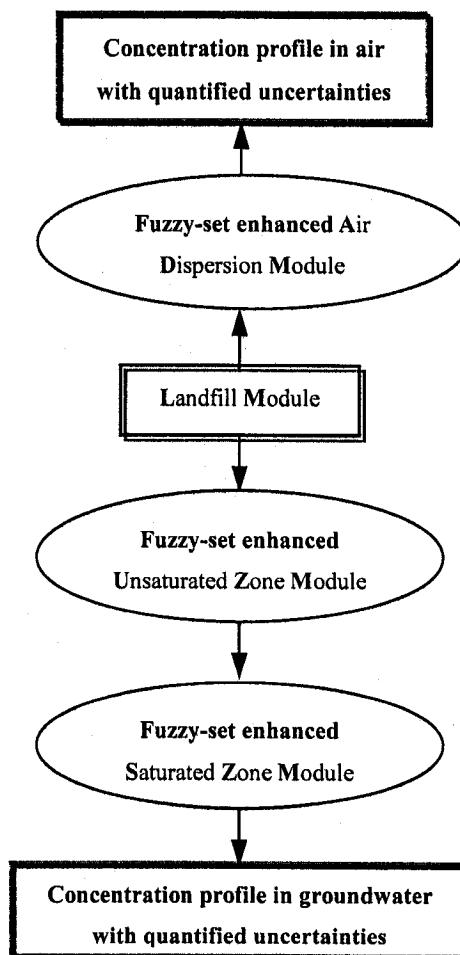


Figure 3.6 A fuzzy-set enhanced EMMS

As shown in Figure 3.6, the landfill module is the core of EMMS comprising a

great deal of uncertainties. The uncertainties are transferred to the linked modules, which are on the transport pathway. These modules are the air dispersion module and the unsaturated zone module. Thereby, the uncertainties in the air dispersion module and the unsaturated zone module arise not only from the module itself but also from landfill module. In this context, the fuzzy set application will be applied in using the following scheme.

1. To analyse the gas dispersion pathway, the gaseous emission flux is the starting point. The emission flux, Equation [3.23], can be transformed into the form ( $M \cdot \text{Transfer function}$ ), in which  $M$  is a key parameter (e.g. the background concentration) involving uncertainty and imprecise information; the transfer function (TF) is a combination of the rest of the equation, which may also contain a number of uncertainties related to pertaining parameters.  $M$  and  $TF$  can be treated as fuzzy numbers, and then the output with quantified uncertainties can be obtained by using the multiplication operation and is then input into the next equation.

Similarly, Equation [3.53] can be converted to the form  $M \cdot TF$ , in which  $M$  is the fuzzy output of the previous equation, and the same fuzzy operation can be performed. It is noticeable that  $TF$  can be regarded as either a fuzzy number or a crisp number, depending on the available data. If it is a combination of various data embracing uncertainties, it is better to treat it as a fuzzy number. Otherwise it can be a crisp number. When  $TF$  is regarded as a fuzzy number or a crisp number, Operation 2 or 3 is used.

Eventually, the pollutant concentration profile in the air can be obtained with the

related uncertainties being quantified.

2. In the leachate transport pathway, fuzzy set application is first applied in the unsaturated zone module, in which the initial contaminant release rate reflects the uncertainties induced by the landfill module. As in the calculation of the gaseous dispersion pathway, the equation for leachate flux computation (e.g. Equation [3.43]) can be replaced by the form  $M*TF$ , using fuzzy operations according to the related characteristics of  $TF$ . By transferring to the following module (e.g. Equations [3.46] and [3.48]), the concentration profile in the groundwater can be calculated with the uncertainties being quantified.

Only the systematic application procedure is demonstrated above. More details can be found and explained in Chapter 4, 5, and 6.

### 3.4 Summary

A fuzzy-set enhanced EMMS has been developed in this chapter with four key modules: a landfill module, an unsaturated zone module, a saturated zone module, and an air dispersion module. The modelling concepts and the expected outcomes are summarized in Figure 3.7.

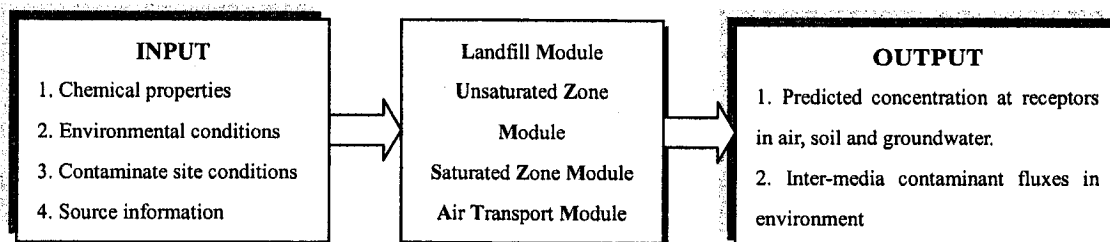


Figure 3.7 The application process of the developed EMMS

The developed EMMS is composed of a set of the state-of-the-art modules, which are capable of the following [1] simulating the internal dynamics of the polluting source area (e.g. landfill); [2] computing emission fluxes up to the air and atmosphere and down to soil and groundwater from the polluting source zone; [3] modelling the subsequent dispersion of pollutants in the ambient atmosphere and modelling the concentration profiles in the connected soil and groundwater systems; [4] not only the physical transport but also the decay evolution have been considered in multiple dimensions; [5] effective uncertainty analysis methods have been developed and integrated within the EMMS to systematically quantify uncertainties associated with key modules and parameters as well as with the multimedia environmental system.

The development of a fuzzy-set enhanced EMMS is provided in this chapter. Two solid model validations I and II follow in Chapters 4 and 5. Chapter 4 includes a systematic testing of the developed EMMS through a complete case study and through a series of modelling comparisons with literature data and studies. Chapter 5 introduces a large-scale field validation based on the Trail Road Landfill in the Ottawa-Carleton Region.

## **Chapter 4**

# **Model Validation I – A Systematic Model Testing**

When the model development is complete, the composite module and the entire modelling system need to be tested and validated to ensure that the model can simulate natural phenomena with reasonable accuracy (van der Heijde, 1987). Thus, a two-step validation process is designed as shown in Figure 4.1. The first step is to examine the performance of EMMS as it was intended to be designed and to ensure that the whole modelling system is fully functional. The second step is to determine how well the mathematical implementation of the natural processes describes natural behaviour of the contaminant in the environment. This determination is based on the degree of correlation between model simulations and actual observed data.

In this chapter, the developed EMMS is first tested in a complete case study; then verification modelling is conducted to compare simulation results with extensively researched literature data; finally, the data from an experimental landfill site is used for a pilot-scale validation. Further field-scale validation is conducted in Chapter 5. The

validation procedure is shown in Figure 4.1.

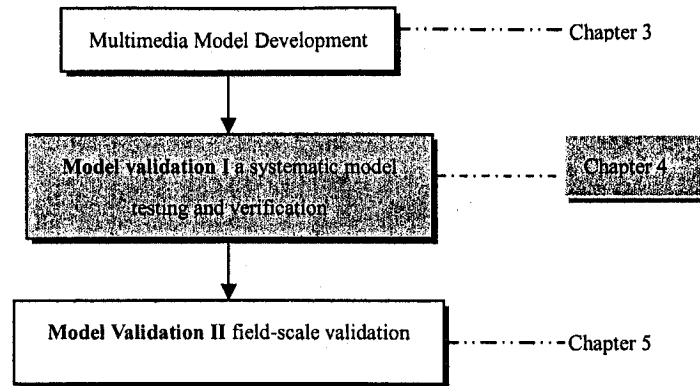


Figure 4.1 The procedure of the model validation

#### 4.1 A Scheme for the Validation of the Developed System

Figure 4.2 conceptually illustrates the scheme of Part-1 validation. A complete case is designed based on a survey of similar cases in the literature. The properties of contaminant, the environmental conditions, and the physical properties of a landfill site are adapted from real available contamination sites in order to use practical data to test the developed EMMS.

The computed results from EMMS is carefully analysed with respect to the reasonable chemical distribution in environmental media and with respect to the physical dynamics of the pollutant in a multimedia environmental system.

Figure 4.3 shows how to organize the validation through comparing the results

of EMMS with the literature data. The surveyed data from the literature is used as inputs to the developed EMMS. As it is impossible to find a complete set of data for the EMMS, key model components are verified using the available data individually. If the simulation results are identical to or close to those from the reference model, the validation is done; otherwise, the model is improved until the comparison error can be accepted.

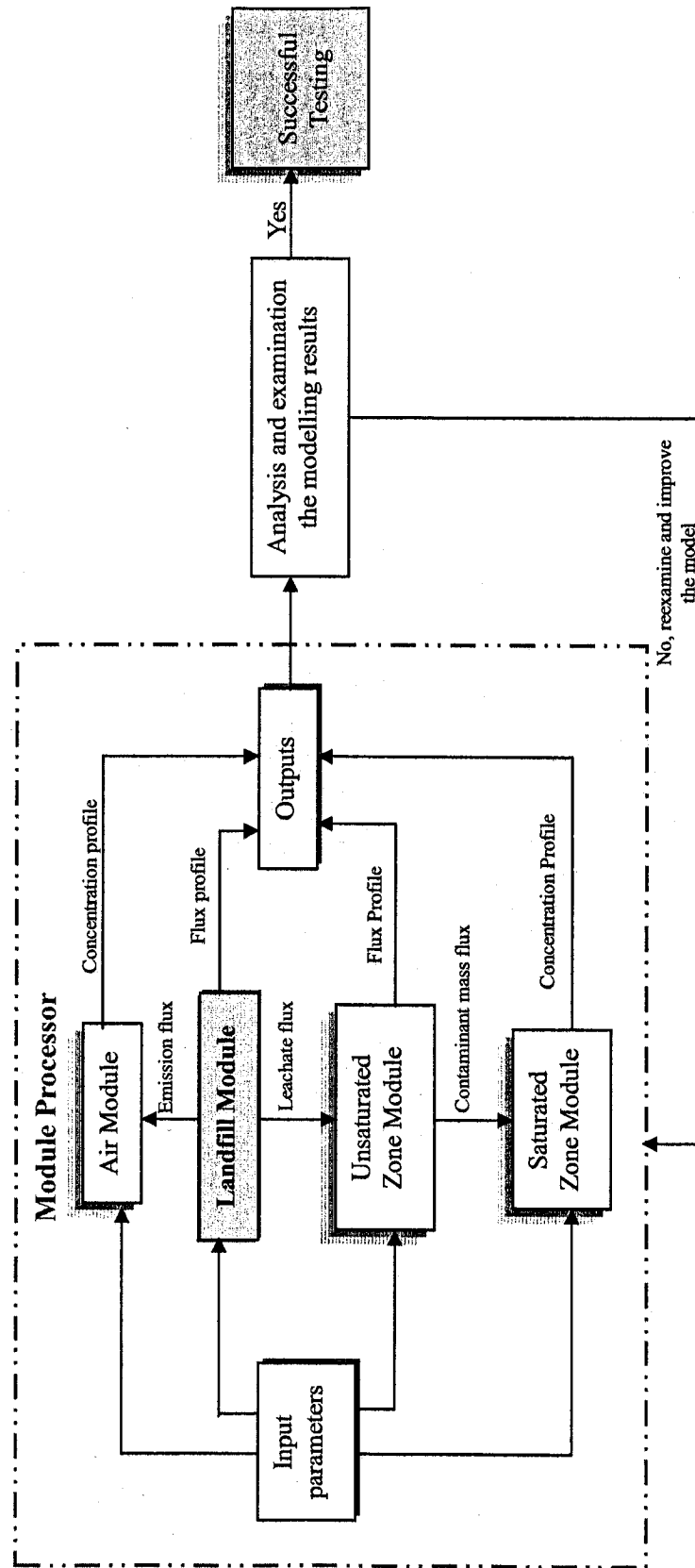


Figure 4.2 The testing of the development EMMS



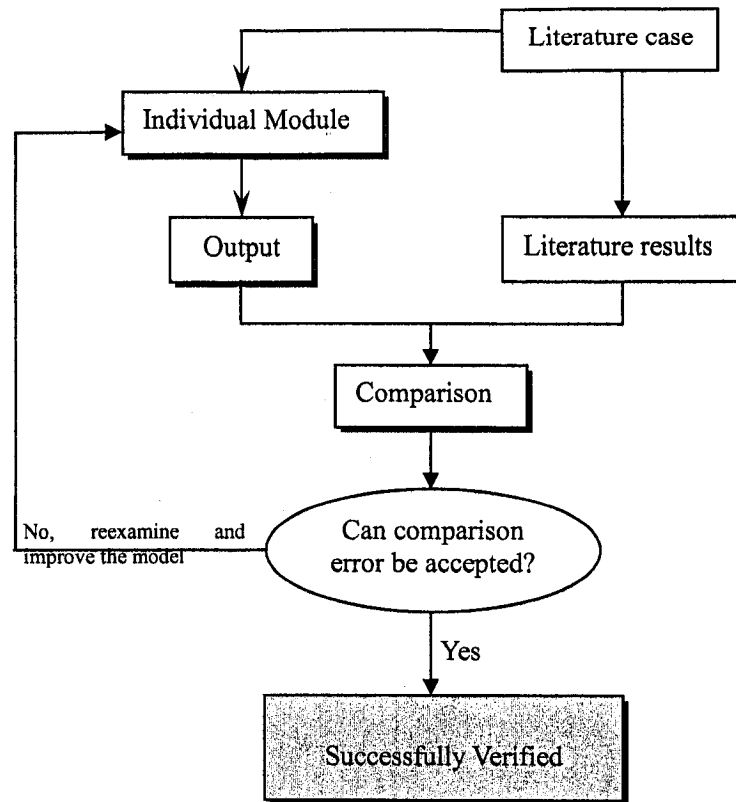


Figure 4.3 The testing of the development EMMS for the accurate performance of modules

## 4.2 Testing of the Developed Modelling System Through a Complete Case Study

### 4.2.1 Overview of the Study System

A landfill site (Rickabaugh 1990, Lin and Hildemann 1995, Labieniec 1996a and 1996b) is used to test the overall performance of EMMS. The site, with an area of  $100m \times 100m$  and a depth of 1 meter, is surrounded by an area of farm, forest, and industry. Moreover, a residential area is located 500 meters downstream from the landfill; and there is a

groundwater pump well located 50 meters downstream from the landfill site and 80 meters orthogonally to the groundwater flow (see Figure 4.4).

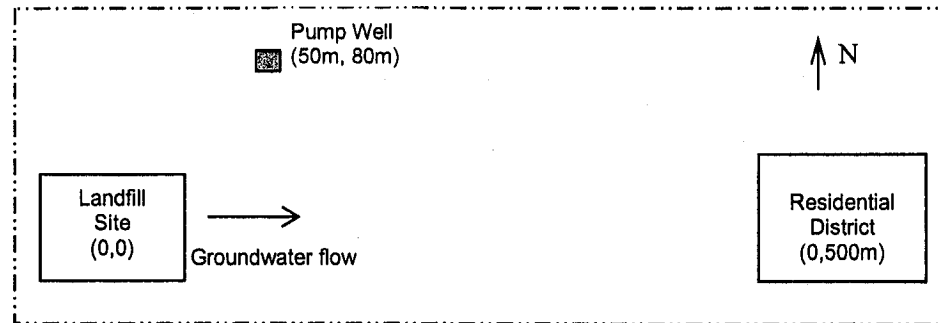


Figure 4.4 The plan of the study site

A polyethylene cover that is 0.05 meter thick is constructed to control the volatile gas emission, but no liner system is installed in the landfill base. A total of 13,500 tonnes of municipal solid waste is loaded into the landfill and compacted to a density of  $1350 \text{ m}^3/\text{kg}$ . Potentially degradable materials include paper, food wastes, plastics, textiles, garden wastes and wood. The composition of the waste is in the typical range shown in Table 4.1.

Table 4.1 Typical ranges of municipal solid waste components

Component		Typical Ranges (%)
Paper		20-65
Glass		2-20
Metals	Ferrous	1-12
	Aluminium	0.5-5
Plastics		2-10
Textiles, rubber, leather		2-15
Organics		10-30
Inorganics and Fines		1-10
Other		0.5-12

Source: (Rickabaugh 1990)

The landfill is located in a city where the precipitation rate leads to  $0.005 \text{ m/d}$  of

bulk leachate velocity in the landfill chamber. A higher leachate velocity has been reported in the literature (Lin and Hildemann 1995). The local meteorological conditions put the area in a stability class of D. The annual surface wind speed is 4.3 m/s.

The soil beneath the landfill site is composed of sand, clay, and gravel; thus the density of the unsaturated zone and the saturated zone is  $1590 \text{ m}^3/\text{kg}$ .

The groundwater level is 3.5 meters under the landfill bottom with a velocity of 0.03 m/d flowing underneath the landfill in a eastern direction. The porosity of the soil and the groundwater near the landfill site is 0.4.

Benzene is chosen to be an investigated contaminant due to its harmful to human health. The initial concentration of benzene in the landfill is around  $1 \mu\text{g}/\text{g}$ .

#### **4.2.2 Modelling Inputs**

The properties of benzene, such as its diffusion coefficients in air and water, its partition coefficient, its half-life of biodegradation, and its Henry's law constant, can be found in the literature (Cohen and Ryan 1989, Jury 1990), and in the chemical database (HSDB 2005). The values of chemical properties of benzene for the studied case are given in Table 4.2. The properties of the soil in the landfill are assumed to be homogeneous with a volumetric air content of 0.2 and a volumetric water content of 0.3. The fraction of organic carbon in the landfill is 0.0125 (Lin and Hildemann 1995). Other input parameters, including the diffusion coefficients of benzene in the unsaturated zone and the saturated zone, are estimated according to the literature (Domenico 1987, and Carnahan and Remer 1984). They are given in Table 4.2-4.5.

Table 4.2 Input parameters of landfill module

the gaseous diffusion coefficient in air	$D_g^a$ (m <sup>2</sup> /d)	0.752	Length orthogonal to groundwater flow	$A_y$ (m)	100
Organic carbon partition coefficient	$K_{oc}$ (m <sup>3</sup> /kg)	$8.2 \times 10^{-2}$	Length parallel to groundwater flow	$A_x$ (m)	100
Henry's law constant (dimensionless)	$K_H$	0.22	The volumetric air content of the soil	$\alpha$ (dimensionless)	0.2
Half-life	$\tau$ (d)	365	The volumetric water content at field capacity	$\theta$ (dimensionless)	0.3
Organic carbon fraction	$f_{oc}$ (dimensionless)	0.0125	Bulk density	$\rho_b$ (kg/m <sup>3</sup> )	1350
The liquid diffusion coefficient in water	$D_l^w$ (m <sup>2</sup> /d)	$8.81 \times 10^{-5}$	Landfill depth	$L$ (m)	1
Liquid velocity	$v_L$ (m/d)	0.005	The thickness of cover	$d$ (m)	0.05
Gaseous velocity	$v_G$ (m/d)	0.005			

Table 4.3 Input parameters of unsaturated zone module

Coefficient of longitudinal dispersion	$D_L$ (m <sup>2</sup> /d)	0.0192
Coefficient of transverse dispersion	$D_T$ (m <sup>2</sup> /d)	0.0027
Average velocity of fluid	$v$ (m/d)	0.005
Porosity	$\phi_{un}$	0.4
Bulk density of unsaturated zone	$\rho_{unSAT}$ (kg/m <sup>3</sup> )	1590
Half-life in unsaturated zone	$\tau_{unSAT}$ (d)	365
Water table depth	$z_{wt}$ (m)	3.5

Table 4.4 Input parameters of saturated zone module

Darcy velocity	$V_d$ (m/d)	0.03
Bulk density	$\rho_{sat}$ (kg/m <sup>3</sup> )	1590
Porosity	$\phi_s$ (dimensionless)	0.4
Organic carbon fraction	$f_{ocSAT}$ (dimensionless)	0.0125
Half-life	$\tau_{sat}$ (d)	365
Dispersivity in x direction	$a_x$ (m)	3
Dispersivity in y direction	$a_y$ (m)	0.3
Dispersivity in z direction	$a_z$ (m)	0.03

Table 4.5 Input parameters of air module

Annual wind speed frequency	$f(\phi)$	0.13
Annual wind speed	w (m/s)	4.3

### 4.2.3 Modelling Results

As conceptually expressed in Figure 4.2, the internal EMMS simulation undergoes the following steps. First, all input parameters are entered in the system; sequentially, the modelling system runs in a designed order to read the corresponding parameters into the landfill module, the unsaturated zone module, and the saturated zone module; finally, once the calculated results are output from the modelling system, they are stored in the categorized output files, including the landfill file, the unsaturated zone file, the saturated zone file, and the air dispersion file. Here, the computation results are shown in Table 4.6.

Table 4.6 Outputs of modelling results

Evaluation time (d)	Concentration (g/m <sup>3</sup> )	Aqueous phase diffusive flux (g·m <sup>-2</sup> ·d <sup>-1</sup> )	Aqueous and/or gaseous phase advective flux (g·m <sup>-2</sup> ·d <sup>-1</sup> )	Gaseous phase diffusive flux (g·m <sup>-2</sup> ·d <sup>-1</sup> )	Total phase flux (g·m <sup>-2</sup> ·d <sup>-1</sup> )
Output at landfill base					
0	0.694	3.29E-5	1.55E-3	0.016	0.0176
365	0.105	0	2.35E-4	0	2.35E-4
Unsaturated zone output					
0	0	0	0		0
365	3.83E-3	9.0E-5	1.91E-5		1.09E-4
Saturated zone output x=50m y=80m					
365	6.15E-017				
Emission Flux from Landfill Cover					
0					2.15E-2
365					1.48E-4
Concentration in air at x=500m					
365	1.85E-10				

At the initial stage of the evaluation period, the concentration of benzene at the

landfill base as shown in Figure 4.5 is  $0.694 \text{ g/m}^3$ . That is also the background concentration that is uniformly distributed in the landfill column. The concentration gradually increases to a peak concentration of about  $0.73 \text{ g/m}^3$  at the early stage. After that peak point, it decreases sharply. Finally, it reaches  $0.105 \text{ g/m}^3$  at the end of one year.

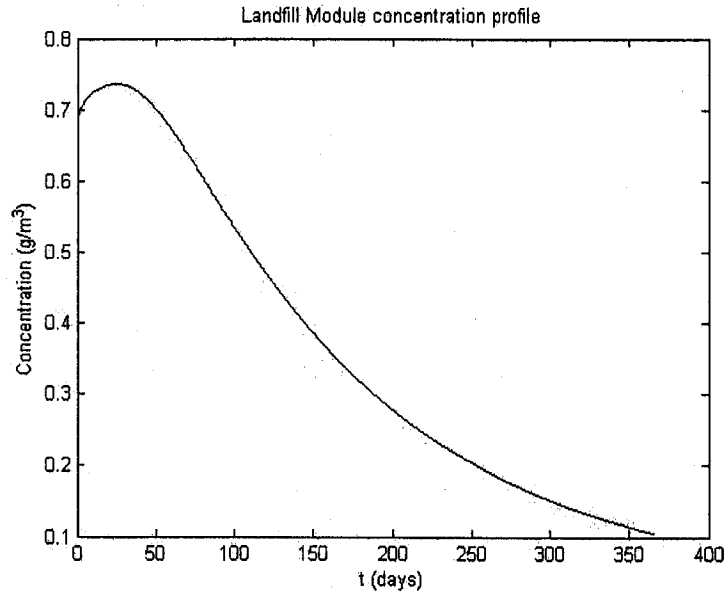
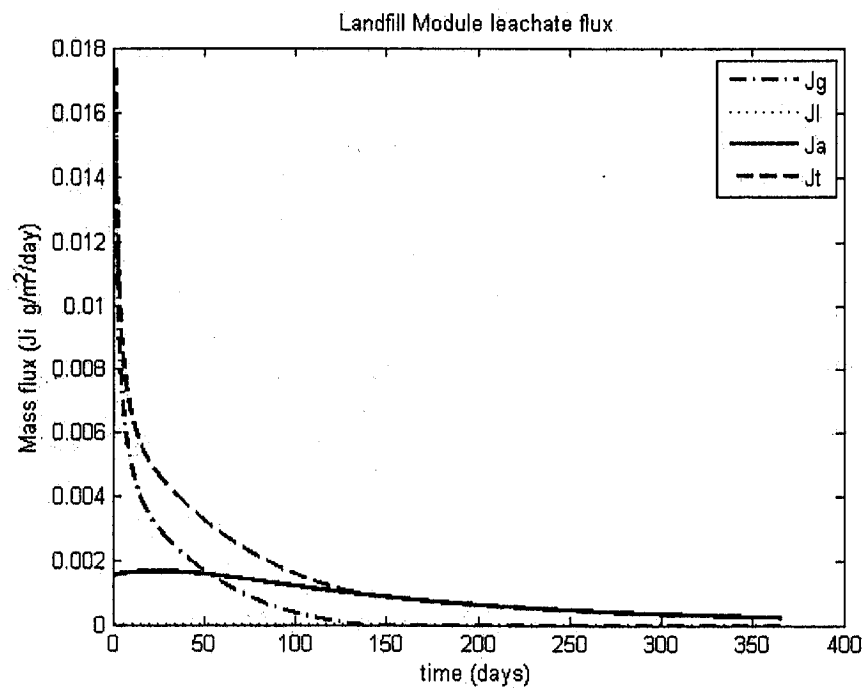


Figure 4.5 The concentration profile at landfill bottom

The total leachate flux draining from the landfill bottom, as given in Figure 4.6, is  $0.0176 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . It is distributed in an uneven pattern into aqueous phase diffusive flux, aqueous and gaseous phase advective flux, and gaseous phase diffusive flux with the value of  $3.29\text{E-}5 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ ,  $1.55\text{E-}3 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , and  $0.016 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ , respectively. It is observed that the leachate released from the landfill bottom line is mainly in the form of gaseous phase diffusive flux and secondarily in the form of aqueous and gaseous phase advective flux. The aqueous diffusive flux has little effect on the leachate leaking due to the small molecular diffusion coefficient in the aqueous phase.

At the end of one year, the total fluxes of leachate moving out of the landfill

have decreased considerably, reaching  $2.35\text{E-}4 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The gaseous diffusion flux has decreased to zero. By contrast, the advective fluxes of the aqueous and gaseous phases that increase slightly at the beginning gradually decrease to  $2.35\text{E-}4 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The diffusive flux in the aqueous phase always remains at a very low level compared to the other fluxes on account of the minute effect contributed by the molecular diffusion coefficient in the aqueous phase, which is 4 orders of magnitude smaller than the gaseous molecular diffusion coefficient.



( $J_g$ : gaseous diffusion flux;  $J_i$ : aqueous diffusion flux;  
 $J_a$ : advective flux;  $J_t$ : total leachate fluxes)

Figure 4.6 The leachate mass flux from landfill bottom

In Figure 4.6, it is seen that the total leachate flux is principally dependent on the varying of diffusive flux in the gaseous phase during the initial 25 days; thus it, like gaseous phase diffusive flux, decreases significantly in this period. In the sequence, the total leachate flux depends on the gaseous and aqueous phase leachate fluxes and

gradually declines over time as those fluxes decrease.

Figure 4.5 and 4.6 show the relationship between the changing concentration profile and the trend of leachate flux over time at the landfill bottom. At the beginning of the contaminant release, the landfill bottom line is the line separating the contaminated soil, which is the mix of soil and landfilled refuse, and the clean unsaturated zone. Thus, the driving force caused by the different concentrations leads to the high leachate flux and the increase of concentration during the first 25 days. As the concentration at the landfill bottom decreases, the difference of concentration, or driving force, between the landfill and the unsaturated zone in the vicinity of the landfill bottom gradually decreases. The leachate flux enters the range of slowly varying.

Figure 4.7 shows the benzene concentration profile at the bottom of the unsaturated zone. The pollutant concentration at the unsaturated zone boundary varies from the initial concentration of  $0 \text{ g/m}^3$  to  $3.83\text{E-}3 \text{ g/m}^3$ ; in other words, the clean soil is contaminated by the leachate flowing downward. Under the conditions in which the pollutant from the landfill is released with an exponential decrease, the concentration front will take relative long time to move downward through the unsaturated zone to the division boundary between the unsaturated zone and the saturated zone. The peak concentration occurs at the end of the year.



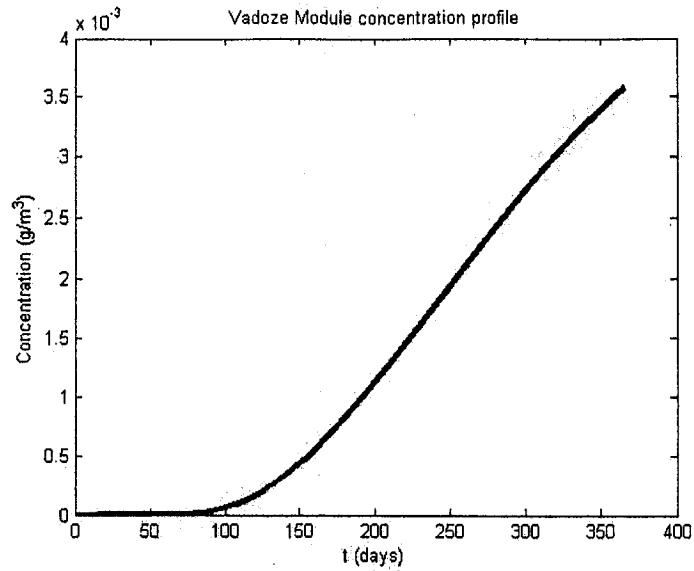
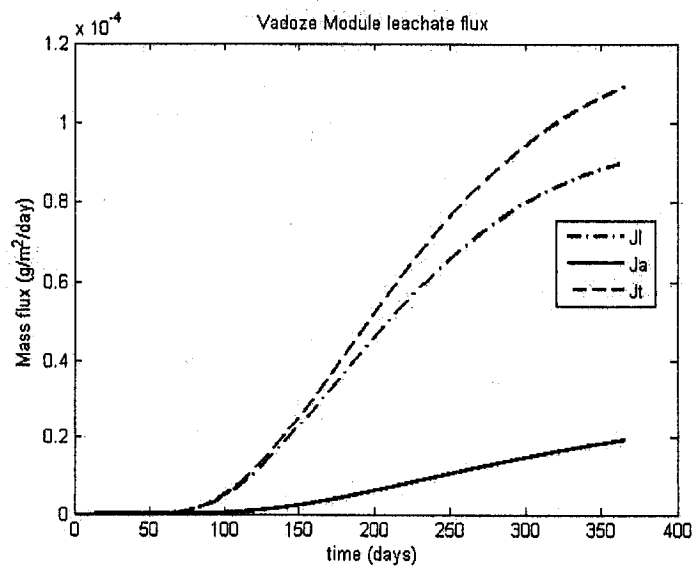


Figure 4.7 The concentration profile at unsaturated zone bottom

At the bottom of the unsaturated zone, the variance of pollutant flux falls into the same trend of concentration for the same reason. The total contaminant flux is gradually from  $0 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  to  $1.09\text{E-}5 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The diffusion flux that accounts for most of the total flux, as shown in Figure 4.8, increases from  $0 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  to  $9.0\text{E-}5 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . The advective flux increases only slightly from  $0 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  to  $1.91\text{E-}4 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ .



( $J_d$ : diffusion flux;  $J_a$ : advective flux;  $J_t$ : total contaminant fluxes)

Figure 4.8 The contaminant mass flux from unsaturated zone bottom

In the unsaturated zone, the contaminant is considered as existing only in a liquid phase and is adsorbed in the solid phase, implying that the diffusion coefficient in the aqueous phase has an effect equivalent to the conjunction of the diffusion in the gaseous and aqueous phases. Therefore, the value of the diffusive flux is estimated to be larger than that in the landfill and much greater than the aqueous advective flux, becoming the leading factor.

One of the final outputs from EMMS is the predicted benzene concentration distribution in the groundwater. Figure 4.9 plots the concentration contour in the groundwater. The predicted concentration at groundwater pump well is  $6.15\text{E-}17 \text{ g/m}^3$  at the end of the evaluation period. The concentration slowly decreases to the downgradient of the groundwater flow, due to the contaminant dispersion and the diffusion in the groundwater.

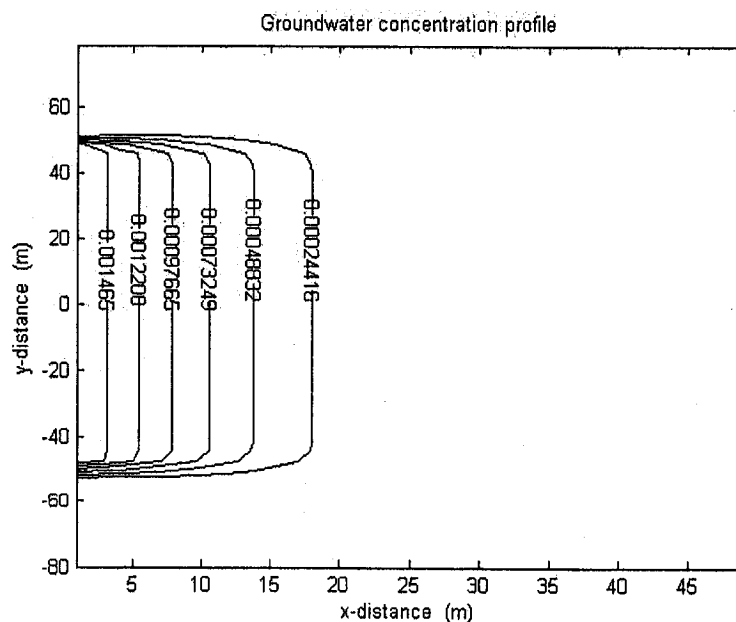


Figure 4.9 Concentration contour in groundwater

The simulations above concentrate on the leachate migration from the landfill to the groundwater pump well. This contaminant migration lead to most concerns about the adverse impacts on human health. Meanwhile, volatile gas emitted from the landfill cover may have the same impact on human health, and cannot be neglected.

Concerning the volatilization of the contaminant from the landfill cover, the benzene emission flux quickly decreases from  $2.15\text{E-}2 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  to  $1.48\text{E-}4 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  during the period of one year, during which the sharply drop of emission flux occurs in approximately 25 days. The varying of the emission flux falls into the same trend of as the leachate flux. The cover on the landfill prevents the contaminant from emitting from the upper side of the landfill. Hence, the majority of benzene is released from the landfill bottom where there is no liner system to block percolation. With the contaminant carried down to the unsaturated zone by the leachate, the emission rate from the landfill cover also quickly dies down as shown in Figure 4.10.

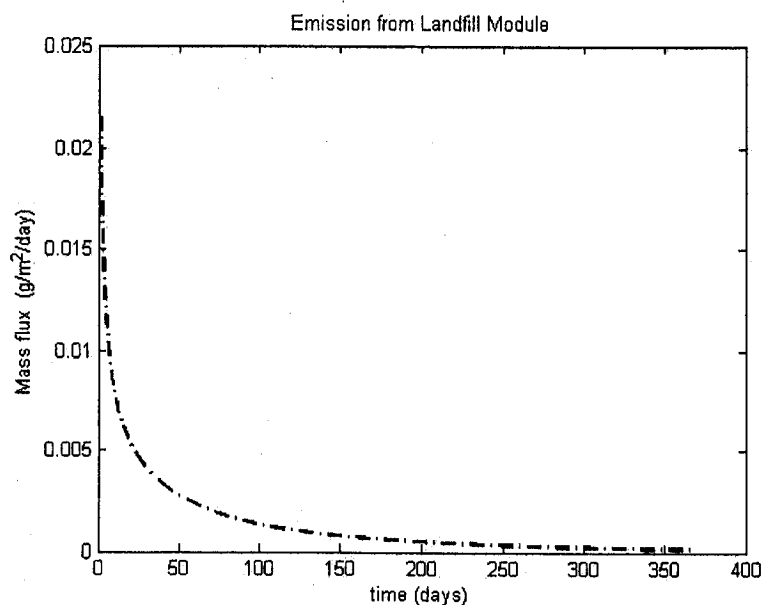


Figure 4.10 Emission flux from landfill cover

With the gas emission significantly decreasing, the concentration of benzene in the air decreases more rapidly along the distance over a short period of time. As a result, the concentration at the receptor that is 500 meters away from the landfill site is so small, reaching a value of  $1.85\text{E-}10 \text{ g/m}^3$ .

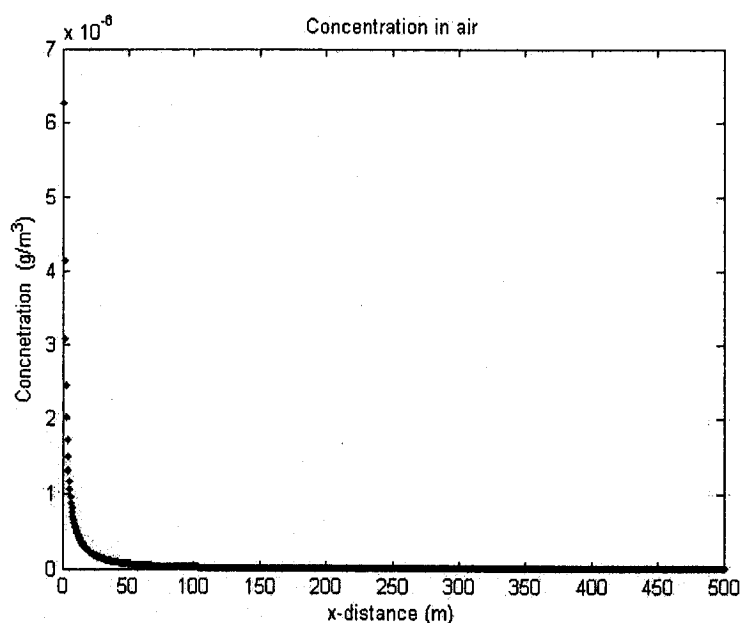


Figure 4.11 Concentration profile in air

Based on an application of EMMS to a complete case study, the simulation results are summarized as follows:

1. Under the study conditions in the landfill chamber, the diffusive gas flux is the predominant mechanism for contaminant release from the landfill. The advection of gas and liquid is the second dominant transport mechanism due to the fact that the effluent directions of the gas and the leachate are opposite. Volatilisation is another dominant loss mechanism, but to what extent it influences the chemical distribution in the gas or liquid

is dependent on the nature of the VOC components.

2. The gas diffusive flux varies substantially with a difference between the contaminant concentration in the landfill and in the unsaturated zone. By comparison, the advective fluxes do not vary significantly. They are related to the velocities of the gas and the leachate and the chemical composition in landfill.

3. In the unsaturated zone, the contaminant gas phase is neglected. Thus, it is the liquid diffusive flux that plays a dominant role in the contaminant transport process, while the liquid advection has minor influence due to the low seepage velocity.

4. A landfill represents one of the most complex emission sources. The release of pollutants from a landfill into the surrounding environment usually increases during the first few years and then decreases. This is due to the decomposition of the solid waste that is buried in the landfill. Accordingly, a time-dependent source term is considered in the EMMS, thereby giving a reasonable modelling output pattern that explains the internal evolution of the solid waste in the landfill as indicated in Figures 4.7 and 4.8.

5. In the saturated zone, the groundwater flow has a higher velocity than the vertical seepage in the unsaturated zone; therefore, the advection is as important as the diffusion in the transport process.

From the analysis above and the examination of the set of modelling results, it can be concluded that the outputs from the connected modules are consistent. In the landfill module, the source module of the multimedia modelling, the emission flux from the landfill cover, the concentration at the bottom of the landfill, and the leachate flux from landfill bottom are reasonably verified with each other. The relationship between the contaminant concentration profiles in the gas phase and the liquid phase is proved

reasonable and is in agreement with the acknowledged characteristic behaviour of benzene in environmental media. Therefore, this environmental multimedia model is reliable and useful for the purpose of environmental risk assessment.

### **4.3 Comparison with Literature Data**

Key module components are applied to the available cases in the literatures, and then the simulation results from EMMS are compared to those calculated from the similar solutions in order to verify that the performance of each module is as good as expected.

#### **4.3.1 Modelling of Landfill Contamination**

The cases of Lin and Hildemann (1995) and Labieniec et al. (1996a) are utilized for the landfill module validation. The first is used to examine the equations used to calculate the concentration distribution in landfill and emission flux from the landfill surface; the other is used to calculate the solution of the leachate flux from the landfill bottom.

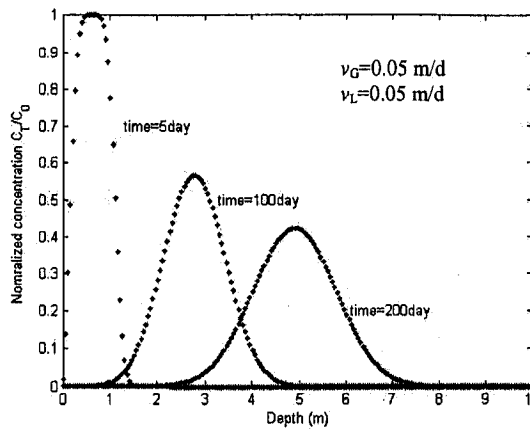
Two compounds, vinyl chloride (VC) and benzene, were chosen to illustrate the simulations for the concentration and emission flux. In the study of Lin and Hildemann (1995), the landfill is assumed to have no landfill cover, and is initially incorporated with uniform contaminants in the uppermost one meter. Table 4.7 lists the chemical properties cited by the authors. Bulk gas velocity  $v_G$  and leachate velocity  $v_L$  are 0.05 m/d and 0.05 m/d for the simulation of the benzene concentration profile in the soil, 0.05 m/d and 0.15 m/d for the VC, and 0.16 m/d and 0.0 m/d for the modelling benzene emission flux

from the landfill cover, respectively.

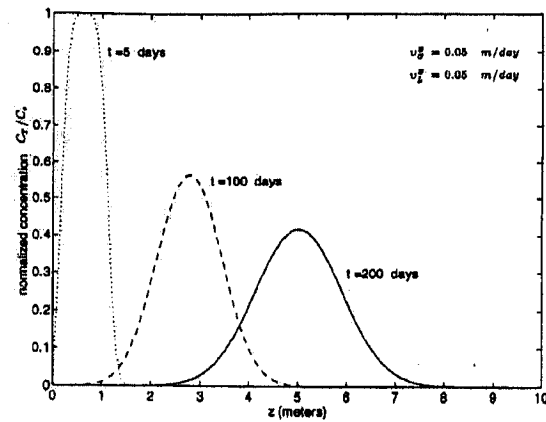
Table 4.7 Chemical properties of two selected compounds (Lin and Hildemann 1995)

Parameters	VC	Benzene
$D_g^a$ (m <sup>2</sup> /d)	0.924	0.752
$D_l^w$ (m <sup>2</sup> /d)	$1.08 \times 10^{-4}$	$8.81 \times 10^{-5}$
$\mu$ (d <sup>-1</sup> )	0	0
$K_{oc}$ (m <sup>3</sup> /kg)	0.4	0.083
$K_H$	97	0.22
$f_{oc}$	0.0125	
$\rho_b$ (kg/m <sup>3</sup> )	1350	
$\alpha$	0.2	
$\theta$	0.3	

Figure 4.12 shows the evolution of benzene in the landfill chamber. The contaminant front moves downwards with peak concentration gradually decreasing with time. After 5 days, the peak concentration appears at the depth of 0.7m with a value of 100% initial concentration ( $C_0$ ); when the time increases to 100 days, the peak concentration is around 55% of  $C_0$  at a depth of 2.5m; and the peak concentration with a value of a little more than 40% of  $C_0$  moves downwards to a depth of 5m after a time period of up to 200 days. As indicated in Figure 4.12, the EMMS modelling results of the benzene evolution match the literature data very well.



EMMS results

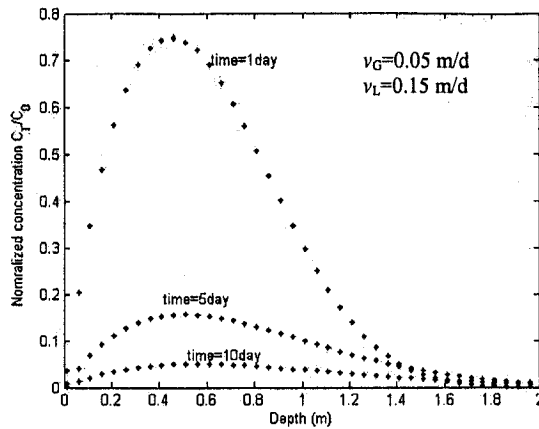


Lin and Hildemann 1995

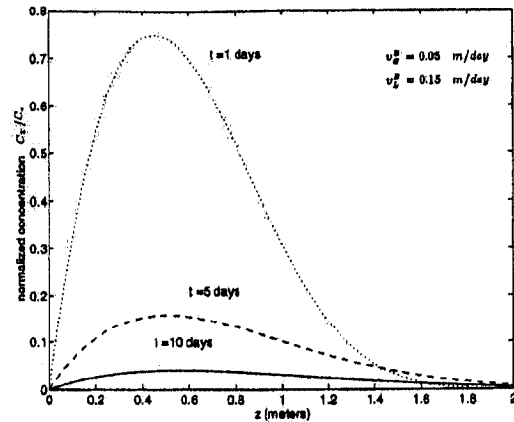
Figure 4.12 Testing of landfill module: the subsurface concentration profile for benzene as function of time

The concentration profiles for VC, a highly volatile compound, are plotted in Figure 4.13. The peak concentration quickly decreases within ten days from around a normalized concentration of 0.75 to 0.03, mainly because the volatility nature of VC results in a higher volatilisation loss to atmosphere than the loss downwards with leachate. Meanwhile, the peak concentrations occur between the depths of 0.4-0.6 meter, meaning that majority of the remaining contaminant is kept in the initially contaminated landfill at a depth of one meter, no migrating with leachate. The peak concentration value and appearing time are also the same as Lin and Hildemann's results (1995) as given in Figure 4.13.





EMMS results

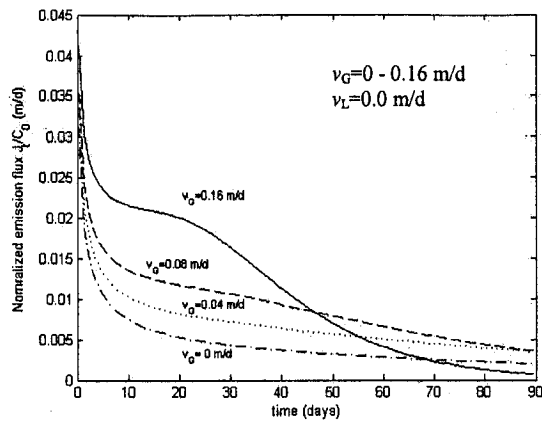


Lin and Hildemann 1995

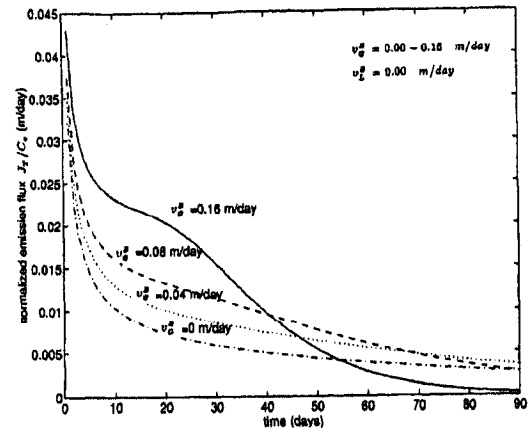
Figure 4.13 Testing of landfill module: the subsurface concentration profile for VC as function of time

The emission flux for benzene is plotted versus time in Figure 4.14. The normalized emission flux decreases from 0.042 m/d to almost zero within 100 days under the effect of a high bulk gaseous velocity of 0.16 m/d and zero bulk leachate velocity. Under this condition, the atmosphere becomes the primary loss pathway for benzene. Also, the emission flux is plotted with the changing of the bulk gas flow in Figure 4.14. The higher the gas velocity, the more the contaminant is swept out from the landfill cover, and the higher emission flux occurs in the early stage.

From the comparison shown in Figure 4.14, the calculated normalized emission fluxes match the data in Lin and Hildemann (1995).



EMMS results



Lin and Hildemann 1995

Figure 4.14 Testing of landfill module: the normalized atmospheric emission rate of benzene as function of time

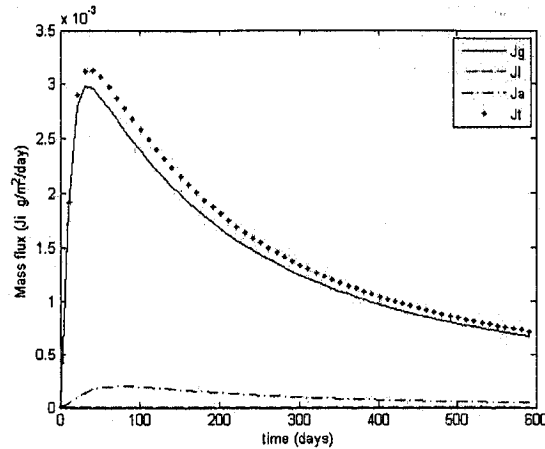
For the calculation of leachate concentration and leachate flux, the study of Lin and Hildemann (1995) did not contain the related data, thus we use another case study by Labieniec et al. (1996a) to verify the model performance. The model used by Labieniec et al. (1996a, 1996b) is based on Jury's model (1983, 1990) and similar to the model of Lin and Hildemann (1995). Thus, this is why the data in Labieniec et al. (1996a, 1996b) can be used to conduct the validation test of the developed EMMS. The selected contaminant is TCE that is initially distributed uniformly in the soil, which is equivalent to the landfill without the cover and without the liner system, with a background concentration of  $1\mu\text{g/g}$ . The required input data are given in Table 4.8

Table 4.8 Input parameters for the computation of leachate flux from landfill (Labieniec et al. 1996a ,1996b)

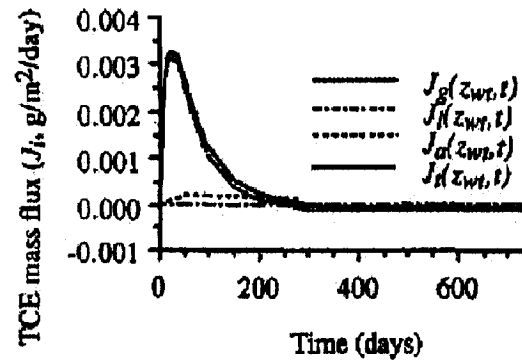
The gaseous diffusion coefficient in air	$D_g^a$ (m <sup>2</sup> /d)	0.785	The liquid diffusion coefficient in water	$D_l^w$ (m <sup>2</sup> /d)	$9.35 \times 10^{-6}$
Organic carbon partition coefficient	$K_{oc}$ (m <sup>3</sup> /kg)	0.124	Porosity	$\phi$	0.3
Henry's law constant, dimensionless	$K_H$	0.41	Bulk density	$\rho_b$ (kg/m <sup>3</sup> )	1590
Half-life	$\tau$ (d)	$4.7 \times 10^8$	Landfill depth	L (m)	1
Organic carbon fraction	$f_{oc}$	0.0075	Gaseous velocity	$v_g$ (m/d)	0.0
Liquid velocity	$v_L$ (m/d)	0.001	The evaluated depth	$z_{wt}$ (m)	1.5

The gaseous and liquid diffusion coefficients are not given in Labieniec et al. (1996a, 1996b). Based on the methods given by Labieniec (1994), these values are calculated and expressed in Table 4.8. Also, some other parameters, such as the volumetric air content, and the porosity of the initially contaminated zone, are not given in the previously studied case. They are estimated based on the same study case and they are also given in Table 4.8.

The mass flux at an evaluated depth over time due to advection and diffusion are presented in Figure 4.15. The peak leachate flux over  $0.003 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  at the evaluated depth occurs after 50 days and then significantly decreases within the next 150 days. It finally remains at almost  $0.0005 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ . These results are closely matched with the literature results despite a small discrepancy as indicated in Figure 4.15. TCE mass flux is zero after 200 days in the results of Labieniec (1996a), but is  $0.0005 \text{ g}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$  in our simulation. This minor difference might be due to the difference in some minor input data used in the two studies.



EMMS results



Labieniec 1996a

Figure 4.15 Testing of landfill module: TCE mass fluxes at water table over time

### 4.3.2 Soil Contamination

The case studied by Carnahan and Remer (1984) is employed in our unsaturated zone module verification. A pollutant is time-dependently and is released continuously into the soil at a rate of 1 mol/yr during 10 years of evaluation time. The required input data are listed in Table 4.9

Table 4.9 Input parameters used in unsaturated zone module (Carnahan and Remer1984)

Coefficient of longitudinal dispersion	$D_L$ (m <sup>2</sup> /yr)	7
Coefficient of transverse dispersion	$D_T$ (m <sup>2</sup> /yr)	2
Average velocity of fluid	$v$ (m/yr)	1
Porosity	$\Phi_{un}$	0.2
Bulk density of unsaturated zone	$\rho_{unsat}$ (kg/m <sup>3</sup> )	1590
Half-life in unsaturated zone	$\tau_{unsat}$ (d)	0
Linear adsorption coefficient between liquid and solid phase	$K_d$ (dimensionless)	10

Figure 4.16 expresses the contaminant concentration profile over the depth. It is clear that after 10 years, due to the retardation of soil sorption, the contaminant concentration movement front has sharply decreased from zero at the releasing source,  $x=y=z=0$ , to around  $10^{-20}$  mol/liter at  $z=16$ m, and  $x=y=0$ , a value which is close to the value given by a similar model developed by Carnahan and Remer (1984).

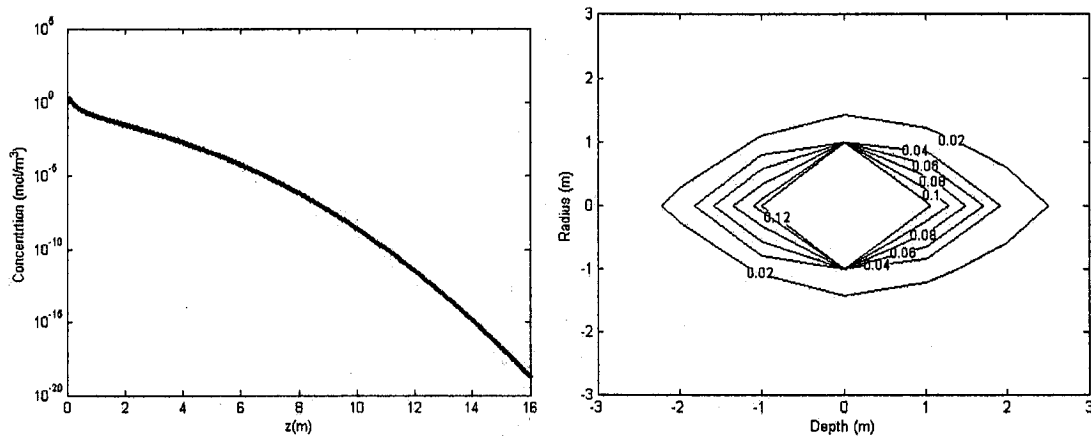


Figure 4. 16 Testing of unsaturated zone module: concentration profile and contour in unsaturated zone.

The concentration contour is plotted in Figure 4.16. With the influence of the flow in  $z$  direction (the depth of soil zone under study), and the different effects contributed by longitudinal and transverse diffusion, the shape of the concentration contour is not circular as shown in Figure 4.16. The concentration near the point of release, at  $r=2$ m, and  $z=0$ m, is  $2.3\text{E-}5$  mole/litre, which is the same as that calculated by Carnahan and Remer (1984).

### 4.3.3 Groundwater Contamination

The case used for testing the saturated zone module is adopted from Domenico (1987).

The contaminant is uniformly distributed in an area source orthogonal to the groundwater flow direction at the initial concentration of  $0.1 \text{ kg/m}^3$ . The dimension of the area source is  $3\text{m}(y) \times 2\text{m}(z)$ , which is orthogonal to the groundwater flow direction. The required input data are presented in Table 4.10.

Table 4.10 Input parameters for saturated zone module (Domenico 1987)

Darcy velocity	$V_d (\text{m/d})$	0.4
Bulk density	$\rho_{\text{sat}} (\text{kg/m}^3)$	1590
Porosity	$\phi_s$	0.4
Half-life	$\tau_{\text{sat}} (\text{d})$	0.01
Dispersivity in x direction	$a_x (\text{m})$	3
Dispersivity in y direction	$a_y (\text{m})$	0.3
Dispersivity in z direction	$a_z (\text{m})$	0.03

The simulation time is 3650 days along the  $y=0, z=0$ , which yields the following results: at  $x=10\text{m}$ ,  $C(10, 0, 0, 3650)=36 \text{ mg/L}$ ; at  $x=100\text{m}$ ,  $C(100, 0, 0, 3650)=0.87 \text{ mg/L}$ . These values are the same as those in Domenico (1987).

Figure 4.17 shows the concentration contour and the concentration profile at the time of 365 days, respectively. They both demonstrate that the pollutant concentration gradually decreases as the groundwater flows downstream and disperses in the y-direction. These chemical variations over distance satisfy the physical mechanism of the chemical's fate and transport in groundwater.

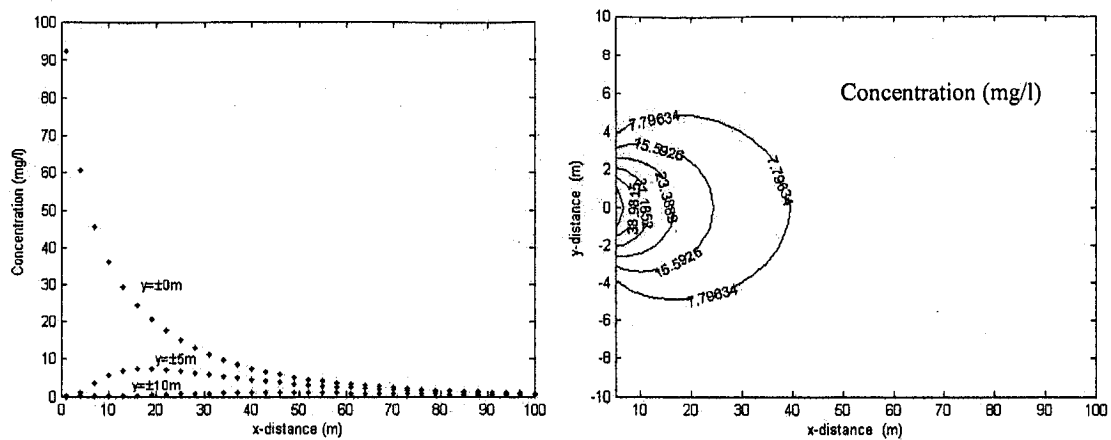


Table 4.11. Following the sorting, the waste was shredded and then loaded into landfill cell lift by lift; it was finally compacted to a density of 474 kg/m<sup>3</sup> (wet weight). The prepared spike chemical was placed between the first and the second lift. A water distribution ring was positioned above the refuse surface with gravels underlying the ring for the purpose of simulating the water infiltration. The gas collection was installed on the refuse surface. The structure of the landfill cell is depicted in Figure 4.18.

Table 4.11 Refuse composition (Rickabaugh 1990)

Component	Percent by weight (%)
Paper	51.7
Food waste	7.23
Fines	6.99
Plastic	6.79
Glass	5.96
Ferrous metal	5.15
Textiles	4.74
Garden waste	3.66
Diapers	2.23
Non-ferrous metal	1.37
Ash-rock-dirt	0.67
Leather-rubber	0.54



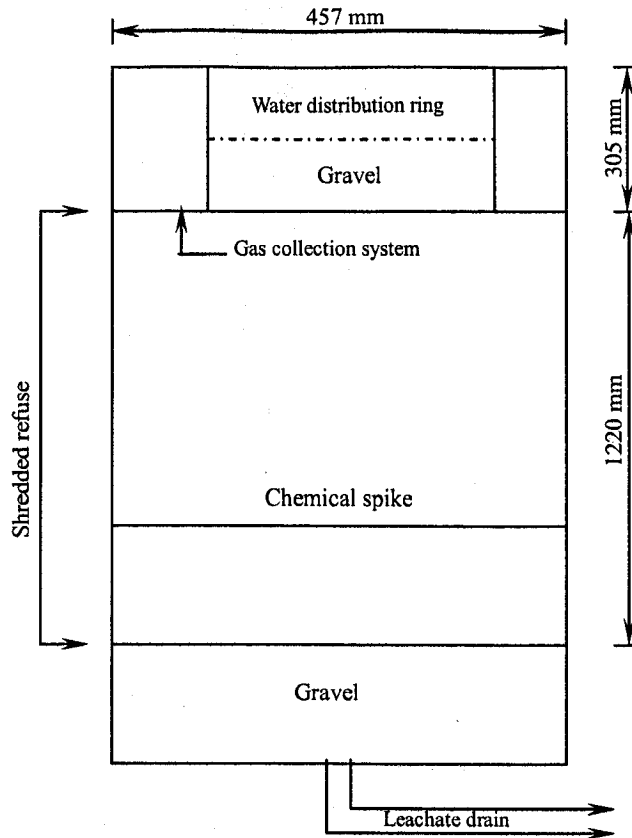


Figure 4.18 The schematic of landfill cell structure (Rickabaugh 1990)

The leachate, gas production, and gas composition were monitored for 5 years. The initial concentration of benzene in the landfill was 83 mg/kg (per kg wet refuse weight). The obtained monitoring data for the benzene emission flux from this landfill cell as reported by Rickabaugh (1990) are used to validate the developed EMMS.

#### 4.4.2 Modelling of Experimental Landfill

The physical properties of experimental landfill and environmental conditions are adapted from Rickabaugh (1990) and given in Table 4.12. The chemical property of benzene is estimated based on empirical methods and is exhibited in Table 4.12.

Table 4.12 Input parameters for modelling the fate and transport of benzene

The gaseous diffusion coefficient in air	$D_g^a$ (m <sup>2</sup> /d)	0.752	Length orthogonal to groundwater flow.	$A_y$ (m)	1.435
Organic carbon partition coefficient	$K_{oc}$ (m <sup>3</sup> /kg)	0.083	Length parallel to groundwater flow	$A_x$ (m)	0.457
Henry's law constant, dimensionless	$K_H$	0.22	The volumetric air content of the soil	$a$	0.15
Half-life	$\tau$ (d)	300	The volumetric water content at field capacity	$\theta$	0.4
Organic carbon fraction	$f_{oc}$	0.0125	Bulk density	$\rho_b$ (kg/m <sup>3</sup> )	474
Cover thickness	$d$ (m)	0.305	Landfill depth	$L$ (m)	1.22
Gaseous velocity	$v_g$ (m/d)	0.0005	Liquid velocity	$v_L$ (m/d)	0.0005
The liquid diffusion coefficient in water	$D_l^w$ (m <sup>2</sup> /d)	$8.81 \times 10^{-5}$			

#### 4.4.3 Simulation Results and Comparison with Literature Data

The simulation period is selected to start from the twelfth month after the refuse was filled into the landfill cell because the gas emission has been tending to stabilize during this time. The simulated benzene emission flux sharply decreases during the initial stage, and then declines smoothly, the value of which show the comparison of the predicted emission flux by using EMMS and the monitored data in Rickabaugh (1990), which shows a high agreement between two profiles as shown in Figure 4.19.

Table 4.13 Modelling results for the pilot-scale landfill

Time (months)	Mean flux (mg·m <sup>-2</sup> ·d <sup>-1</sup> )	Experiment results (mg·m <sup>-2</sup> ·d <sup>-1</sup> )
12	66	61.25
14	21.2	29.34
17	9.1	15.7
19	5.6	4.5
25	1.9	3.3
29	1.7	1.89
31	1.2	1.0
32	0.9	1.1
33	0.9	1.03
34	0.7	0.6

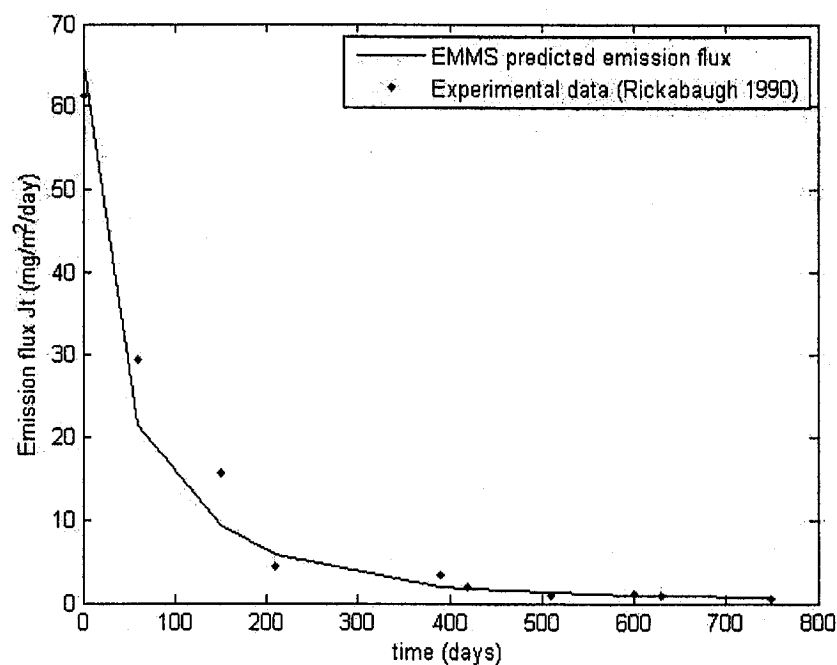


Figure 4.19 Comparison of the emission flux of benzene between the predicted and observed results

Using real experimental data further confirms that the developed EMMS is a reliable modelling tool. These modelling practices also reveal that various uncertainties are associated with the environmental conditions, the release source, the model

formulation, and the model parameters. It is important for the developed EMMS to have an effective method for quantifying these uncertainties.

#### 4.4.4 Quantification of Uncertainties Using a Fuzzy-Set Approach

This subsection is intended to quantify the uncertainties associated with the modelling study in Section 4.4.2. Referring to Section 3.3.2, the detailed steps for incorporating fuzzy methodology are explained below:

- 1). Transform Equation [3.23] into the form

$$J_t(z,t) = C_{L0}TR_e \quad [4.1]$$

in which  $C_{L0}$  is the background concentration in the landfill ( $\text{g/m}^3$ ), and  $TR_e$  is named transfer function in landfill ( $\text{m/d}$ ), which represents the rest of Equation [3.23] and combines the uncertainties in the estimation of input data and in the modelling building. Thus, viewing  $C_{L0}$  and  $TR_e$  as fuzzy numbers is a method for coping with the uncertainties in the landfill module.

- 2). Equation [3.24], the integration of  $J_t$ , can be considered as  $\sum_0^t J_t \Delta t$ , thus the addition operation on the fuzzy numbers can be applied and fuzzy set  $M_{Vol}(t_{ad})$  is produced.

- 3). By using the same method as in Step 1, Equation [3.53] is transferred into the form

$$C_{air} = M_{Vol}(t_{ad})TR_a \quad [4.2]$$

where  $TR_a$  is a transfer function ( $1/\text{m}$ ) combining the uncertainties and imprecision of the wind speed and the vertical dispersion coefficient; thereby it can be viewed as a fuzzy

number to attain prediction intervals and to enlarge the decision space for assessment.

The conceptual fuzzy set approach is illustrated in Figure 4.20.

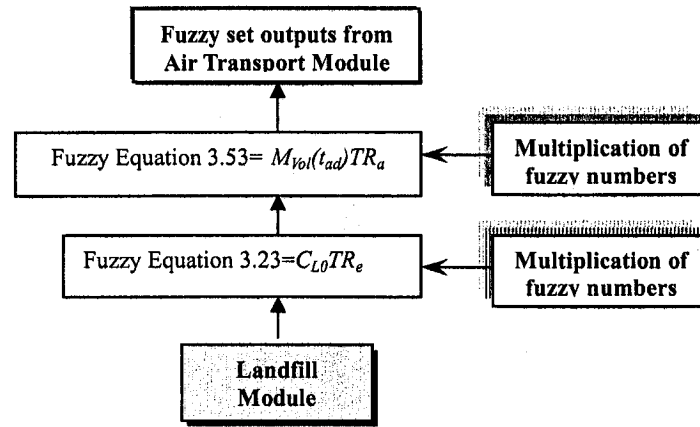


Figure 4.20 Fuzzy analyses for quantification of uncertainties associated with the air pollutant dispersion in the pilot-scale landfill

Applying fuzzy set operations to Equations [4.1] and [4.2], fuzzy set outputs of EMMS are computed. By computing the membership functions of the fuzzy outputs that into which the observed data fall, it is demonstrated how various sources of uncertainties are quantified.

#### 4.4.5 Uncertainty Analysis on Experimental Landfill

Using Equation [3.55] we can calculate the memberships of observed data falling into the interval confidences of fuzzy outputs so that the EMMS's capacity for handling uncertainty is enhanced.

Table 4.14 The results of uncertainty analysis for benzene

Time (months)	Low bound of flux ( $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ )	Mean flux ( $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ )	High bound of flux ( $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ )	Experiment results ( $\text{mg}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ )	The possibility degree
12	39.6	66	92.4	61.25	0.18
14	12.7	21.2	29.7	29.24	0.96
17	5.4	9.1	12.7	15.7	-
19	3.3	5.6	7.8	7.5	0.48
25	1.1	1.9	2.6	3.4	-
29	1.0	1.7	2.3	1.89	0.32
31	0.7	1.2	1.7	1.0	0.40
32	0.6	0.9	1.3	1.1	0.50
33	0.5	0.9	1.2	1.06	0.43
34	0.4	0.7	1.0	0.6	0.33

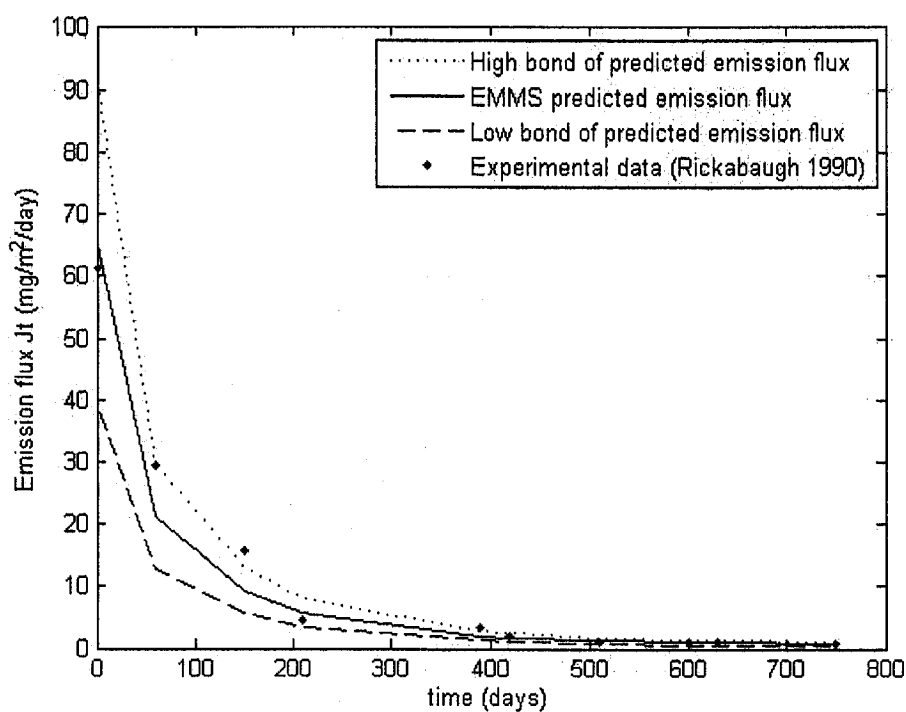


Figure 4.21 Quantification of uncertainties associated with the air pollutant dispersion in the pilot-scale landfill

As seen in Table 4.14, the calculated mean contaminant flux emitted from the

landfill cover is close to the experimental results; as a result, the experimental concentration data fall into a relative high possibility degree of fuzzy outputs. The reason for this high agreement lies mainly in the following aspects:

1. Experiment landfill is only around 1 meter in depth. The estimated chemical properties obtained from laboratory experiments are more suitable for this type landfill.
2. The weather conditions at the experimental landfill site are relatively stable compared to a large-scale landfill field. This also helps achieve a better model validation.
3. The pollutant of concern is prepared and added into the landfill. They are not mainly from the decomposing of refuse. Thus, the emission of chemicals is more steady than that from a field landfill.

## **4.5 Summary**

According to Figure 4.2 and 4.3, EMMS has been validated through a series of case analyses including real experimental data.

This section is summarized as follows:

By applying EMMS to a complete case, the systematic operation and all the functions of the developed EMMS are clearly illustrated and tested.

By examining the modelling results, it is understood that the main processes dominating the fate and transport of chemicals in a landfill chamber and soil are advection, diffusion and volatilisation, but to what extent they influence is dependent on the nature of the waste and the landfill setting.

The second step of validation is to examine the accurate performance of EMMS.

The detailed comparisons between EMMS outputs and the literature data prove that EMMS can perform the accurate modelling simulation. It can therefore be concluded that the modules of EMMS have been correctly developed to support the entire environmental multimedia modelling system.

In addition to the testing of the modelling system performance, EMMS is applied to a pilot-scale landfill site to validate the modelling of volatile organic compounds released from a landfill cover. This application is achieved by comparing the observed contaminant emission flux from the landfill cover with the modelling results. Finally, a fuzzy set approach is integrated into EMMS to enhance its modelling capacity with respect to the quantification of various uncertainties. This integration has also been well examined through the pilot-scale modelling study.



## **Chapter 5**

### **Model Validation II - Field Validation**

The developed EMMS is systematically tested and explained using both literature and real experimental data in Chapter 4. However, the appropriateness of applying the EMMS to a large scale field contamination site is not clear. The Trail Road Sanitary Landfill in the Region of Ottawa-Carleton kindly provided data sets with a large amount of landfill site data (TRNL 2002), which has enabled us to conduct a field validation of the developed EMMS in this chapter.

It is fairly difficult to find an appropriate site from which all the needed validation data can be obtained. Therefore, following the validation of EMMS on an experimental landfill to examine the modelling ability of producing the concentration in air, the observed contaminant concentrations in the groundwater underneath one landfill and in its vicinity are employed to validate the systematic modelling of contaminant leachate flow from the landfill to the groundwater.

Furthermore, the uncertainty analysis is performed with the aim of finding a better way to deal with the uncertainty existing in the modelling system and with the ambiguousness of the observed data from the monitoring program.

## **5.1 Overview of the Trail Road Landfill Site**

### **5.1.1 Location**

The Trail Road Sanitary Landfill site, which includes the Nepean and Trail Road landfills, is located within the Region of Ottawa-Carleton, Canada. It has a population of 750,000. The site, approximately 500 acres, is surrounded by light industry and farmland. The eastern boundary of the site is Highway 416, and the western side is bordered by Moodie Drive. Barnsdale Road runs along the south end of the site, but not immediately adjacent to the landfills. Cambrian Road runs through the northern boundary of the site. The closest residence is 0.85km from the landfill site boundary. The closest residential subdivision is Barrhaven, which is about 2km away (See Figure 5.1 and Figure 5.2).

Approximately 500 meters from the northern boundary of Trail Road Landfill is a large de-watering pond used to catch the local groundwater discharge. Jock River is located approximately 1 km from the north of the pond, the water of which eventually discharges into it. Southwest of Trail Road is the Nepean Landfill.

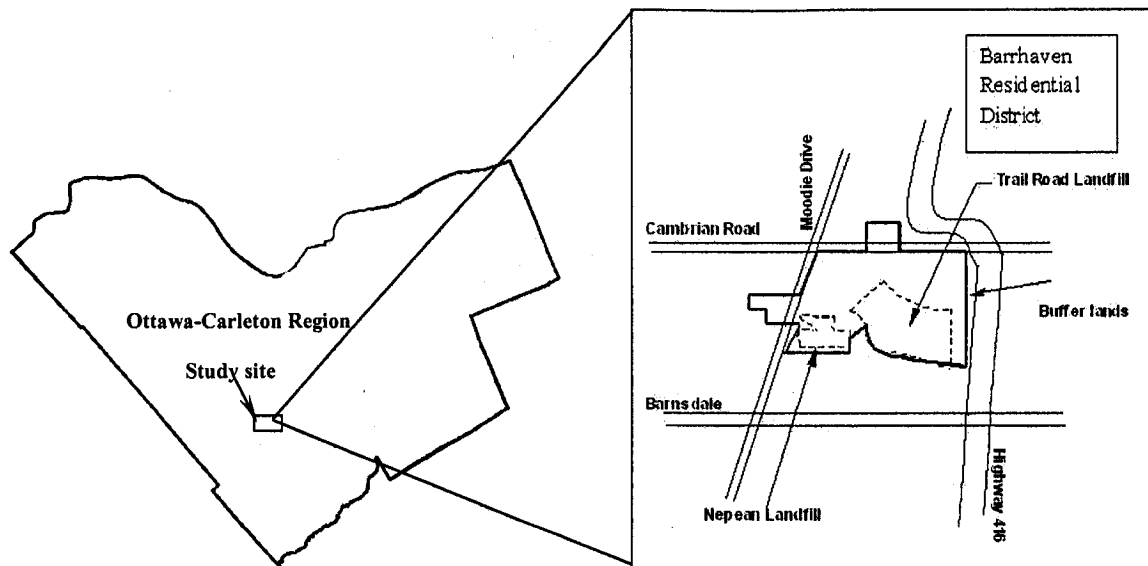


Figure 5.1 The Trail Road Sanitary Landfill

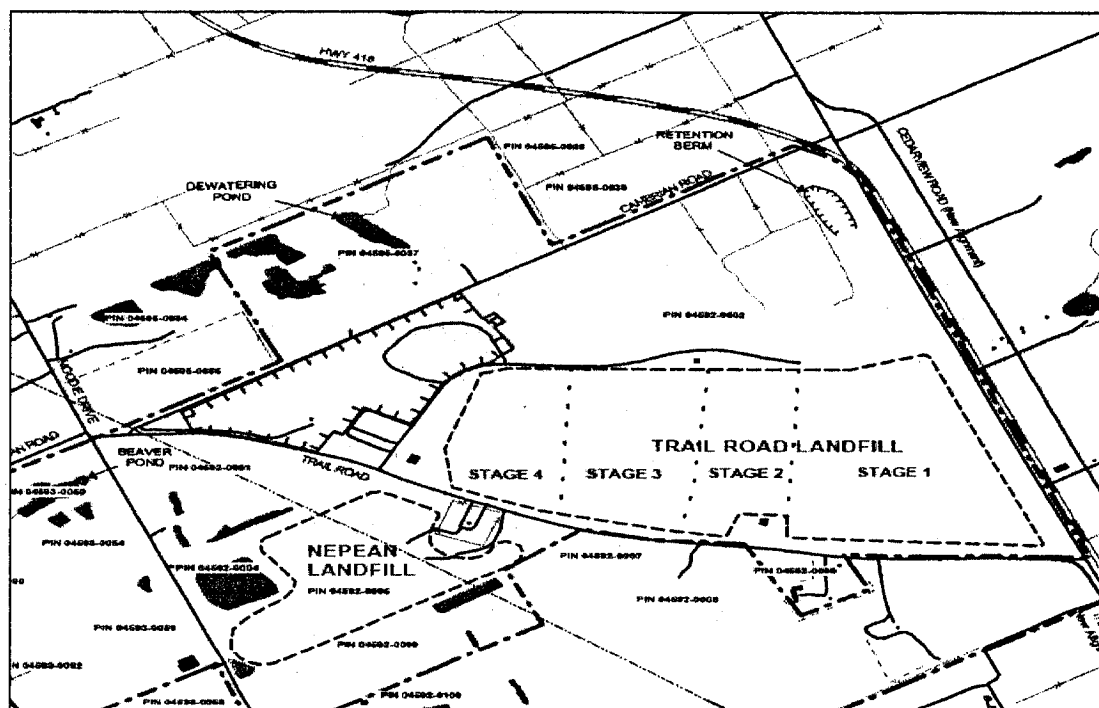


Figure 5.2 Site plan of Trail Road Sanitary Landfill (TRNL 2002)

### 5.1.2 Geological Condition

The landfill site is situated on a glacial outwash plain that has a complex mixture of sands, gravels, cobbles, clays, and silt (Figure 5.3). There are two aquifers, separated by silt and clay, completely underlying the Nepean Landfill but partially under the Trail Road Landfill.

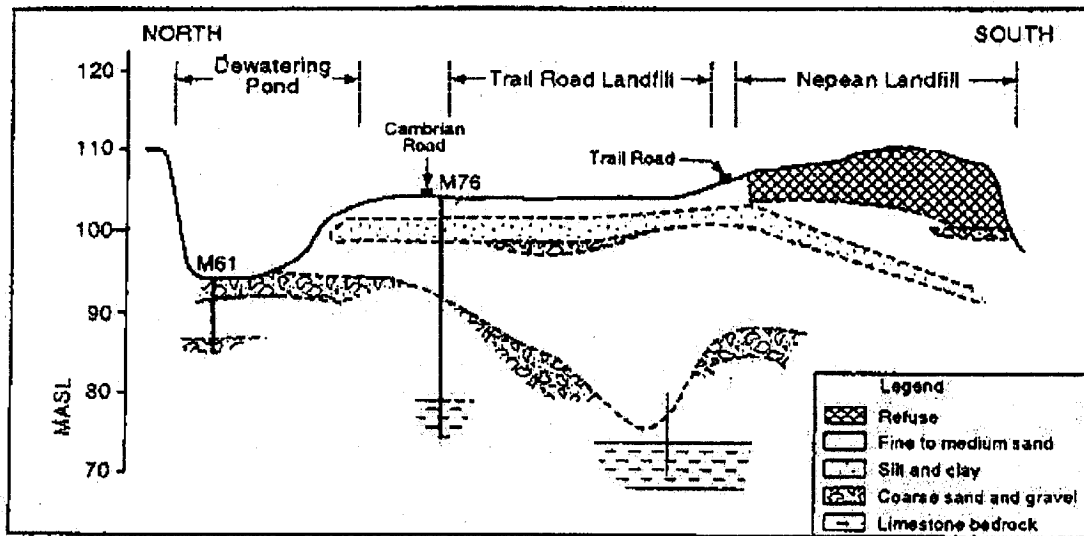


Figure 5.3 North to South Cross Section of Site (TRNL 1995)

A shallow sand aquifer flows in a north-to-northeasterly direction under the Trail Road Landfill. Surface water penetration creates a shallow groundwater flow in a south-to-southwesterly direction under the Nepean Landfill. The deep aquifer, located in a layer of bedrock at a depth ranging from 10-30 meters, flows in a south-to-north direction.

### 5.1.3 Physical Hydrogeology

There are four main hydrostratigraphic units from surface to depth identified beneath the Nepean landfill and partially under of Trail Road landfill:

- the shallow aquifer: comprising the fine to medium sand geological unit;
- the clay aquitard: comprising the marine silt and clay geological unit;
- the deep aquifer: comprising the sand and gravel geological unit; and
- the bedrock aquifer: comprising the dolostone bedrock geological unit.

The shallow aquifer is the upper aquifer beneath the Nepean Landfill, beneath parts of Trail Road Landfill and in the northwest corner of the Trail Road Landfill site.

The typical hydraulic conductivities for the shallow aquifer vary between greater than  $2.0\text{E-}2$  cm/s to a lower limit of  $1.3\text{E-}5$  cm/s. The hydraulic conductivity of the shallow aquifer is variable due to spatial variations in the gradation of the sand and the silt and clay content.

The clay aquitard is composed of the marine silt and clay that underlies the Nepean Landfill as well as north and northeast of the Trail Landfill. The hydraulic conductivities are on the order of  $1.0\text{E-}7$  cm/s for the low permeability clay aquitard.

The deep aquifer lies beneath the Trail Landfill. The groundwater flow direction in the upper/middle part of the deep aquifer is northwest, generally directed to the Dewatering Pond, and is northwards immediately to the south and west of the Trail Landfill.

Groundwater flow is not found in the upper/middle deep aquifer in the vicinity immediately west and southwest of the Nepean Landfill and northeast of the Trail Landfill due to absence of the upper/middle deep aquifer in this area.

Groundwater flow in the lower part of the deep aquifer from beneath most of the Trail Landfill site is towards the northwest. Particularly, the bulk of groundwater from beneath Stages 1 and 2 in the deep aquifer flows to the north and northwest. Beneath the

Nepean Landfill, it is towards the northwest and west. It is not shown south and northeast of the Trail Landfill in the lower deep aquifer due to the small number of monitoring wells in the lower deep aquifer in this area.

Information shows that typical hydraulic conductivities for the sand to sand and gravel aquifer materials vary between greater than  $1.0\text{E-}2$  cm/s to a lower limit of  $1.0\text{E-}04$  cm/s. The hydraulic conductivity is variable in the deep aquifer due to variations in silt, clay and fine sand content.

Horizontal hydraulic gradients in the shallow and confined deep aquifer based on the data for the year 2002 range from negligible to 0.03. Locally, near the edge of the clay in the shallow aquifer, gradients may be much greater.

TRNL (2002) shows that groundwater flow velocities are estimated to range from several metres to several tens of metres per year in the deep aquifer and from several centimetres per year to several metres per year in the shallow aquifer. These estimates are based on the measured hydraulic conductivities and a porosity of 0.3. The vertical hydraulic gradients in April of 2002 between the upper parts of the water table deep aquifer and lower parts of the water table deep aquifer are in the order of 0.008 downward to 0.007 upward.

Where the clay aquitard is present (i.e. in the vicinity of the Nepean Landfill) the vertical hydraulic gradients between the shallow aquifer and the deep aquifer are on the order 0.3 to 0.7 downward (monitoring sites M50 and M51), reflecting the effectiveness of the aquifer separation provided by the low permeability of the clay aquitard. The vertical hydraulic gradients within the confined deep aquifer vary from 0.07 downward near the top of the aquifer to 0.02 upward near the bottom of the aquifer (monitoring sites M50 and M51).

North of the Trail Landfill, the groundwater flow in the shallow aquifer is to the north/northeast and in the confined deep aquifer it is to the northwest.

Horizontal hydraulic gradients in the shallow aquifer and confined deep aquifer based on 2002 data range from negligible to 0.03. The vertical hydraulic gradient in April of 2002 between the shallow and deep aquifers was on the order of 0.02.

There is no contaminant found in the bedrock aquifer, thus this aquifer is not taken into account in this study.

#### 5.1.4 Climatic Condition

The temperature of the Ottawa region where the Trail Road landfill is located significantly changes between summer and winter, and is subject to unpredictable weather conditions. In winter there is generally snow and ice. Temperatures in winter can drop as low as  $-25^{\circ}\text{C}$  at night. Summer weather in Ottawa is warm and humid with temperatures often exceeding  $30^{\circ}\text{C}$ , sometimes as early as April and as late as October. However, summers are usually short, and spring and autumn are unpredictable, with early or late snowfalls possible or even unseasonal heat waves.

The weather statistics displayed here represent the mean value of each meteorological parameter for each month of the year. The sampling period for this data covers 30 years from 1961 to 1990 (Weather Network).

Table 5.1 Climate information of landfill site

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
Rainfall (mm)	58	59	65	69	76	77	88	92	83	75	86	83
Mean Temp ( $^{\circ}\text{C}$ )	-10	-8	-2	6	13	18	21	19	14	8	1	-7
Wind Speed (km/h)	16	16	16	16	14	13	11	11	12	13	15	15
Wind Direction	W	W	W	NW	SW	SW	SW	SW	SW	SW	E	W

## **5.2 Field Validation of the Developed EMMS Based on the Trail Road Landfill Site**

### **5.2.1 Operation History of the Landfill**

The Trail Road Sanitary Landfill has been operated in stages. The first operation landfill was Nepean Landfill, which began operation in the early 1960s and accepted waste until the early 1980s at which time it was deemed nearly full. The Nepean Landfill was capped with a polyethylene liner and soil in 1993.

After the Nepean Landfill closing, the Trail Road Landfill was opened. It is currently serving as a municipal sanitary landfill accepting solid non-hazardous waste including residential garbage and construction, commercial, institutional, and light industrial waste. The Trail Road Landfill was opened in 1980 and has been continuously operated in stages (see Figure 5.2). The first two stages have been closed and capped with polyethylene and soil but are not lined and do not have leachate collection systems. Stage 3 was constructed with 60-centimetre thick competent clay and a high-density polyethylene liner. The third stage, which opened in 1991, is nearly full, and will be capped with a polyethylene liner and soil. Stages 3 and 4 have leachate collection systems (TNRL 2002).

The landfill site is approximately 150 ha in area, of which about 65 ha is the fill area. Stage 1, approximately 25 hectares, was opened in 1980 and finished receiving waste in 1986. Stage 2, about 16 hectares, received waste as of 1986 and was completed in 1991 (TRNL 1995, 2002).



### **5.2.2 Rate of Refuse Disposal**

It is difficult to obtain a detailed annual disposal rate of Nepean Landfill and Trail Landfill for the year of 1960 to the year of 1992 due to the lack of historical operation information. Therefore, based on the data from TRNL (1995, 2002), the rate of refuse annually disposed of as landfill is estimated as 287,000 tonnes/yr.

### **5.2.3 Collection and Estimation of Characteristic Model Data**

Imprecise and inaccurate input parameters are the primary source of modelling error for the environmental risk assessment. Sometimes, data observed can even be wrong due to sampling and analysis errors. For example, so far as the half-life of a chemical is concerned, there may be significant discrepancy between its value measured in the laboratory and the actual value on site. On the other hand, the determination of a parameter value itself carries inherent uncertainty since the processes simulated by the model have a large natural variability in time and space. Moreover, such values are often derived from the experimental data that refer to only a few discrete points in time and space (USEPA 1996).

In this context, a guide is given in this section to select or estimate important parameters that have a relatively great potential influence on the modelling outputs. These parameters may not be suitable for the other sites where environmental conditions are different. In the next four subsections, four categories of model parameters are obtained following the approach outlined by IAEA (1989).

### 5.2.3.1 Properties of the contaminants

Benzene, ethylbenzene, and toluene (BET) are common contaminants of groundwater near landfill sites and have been found to have adverse health effects in toxicology studies (Jury et al. 1990). Thus, BET is chosen as the contaminants of interest in this case study.

#### Half-life and degradation rate

The estimated half-life is the time required for one half of the compound to biodegrade. The estimated half-life is used to calculate the first order rate decay of a chemical using Equation [3.14]. Thus, the estimation of the half-life of a pollutant plays an important role in predicting the concentration in the media of concern.

The biodegradation of BTE occurs both in soil and groundwater. In Howard (1990) and Howard et al. (1991), the degradation rate of BTE ranges from a few hours to hundreds of hours in soil and in groundwater, and it is much shorter in soil than in groundwater. That means, that benzene, as an example, decays from an initial concentration to zero in a few days in soil, but it will take almost two years to complete this process in groundwater. Such a discrepancy between two degradation rates is primarily because the chemical half-life in soil given in the literature usually refers to it in near-surface soil. The chemical degrades faster under aerobic conditions. By comparison, due to anoxic conditions typically found in organic contaminated aquifers, anaerobic degradation of aromatic hydrocarbons plays an important role in the degradation processes occurring in groundwater (Jahn et al. 2005).

As for the soil in a landfill and underlying the landfill, it is obvious that the environment cannot be treated as near-surface soil since the contaminant can be distributed tens of meters beneath the ground surface. Therefore, it is more suitable to consider that in a landfill the degradation of a compound occurs in conditions that are mainly anaerobic, as happens in groundwater.

Hutchins et al. (1991a) evaluated biodegradation of aromatic hydrocarbons under anaerobic conditions, particularly denitrifying conditions, in a series of laboratory tests. First-order rate constants range from  $-0.0224$  to  $-0.035$  for toluene and from  $-0.00174$  to  $-0.00229$  for ethylbenzene. The authors believe that the experimental results are strong presumptive evidence that denitrifying bacteria are responsible for toluene biodegradation. They also found that the biodegradation of toluene did not occur when nitrate is absent from the experiment. In addition, the rate of biodegradation is affected by the pollutant concentration, the temperature of incubation, and the availability of nitrate.

For the benzene biodegradation under denitrifying conditions, Hutchins et al. (1991a, 1991b) thought biodegradations needs more evaluation from the regulatory standpoint due to the fact that its results are totally different from those of the previous study (Bouwer and McCarty 1983, Zeyer et al. 1986, Kuhn et al. 1988, Majora et al. 1988, Berry-Spark et al. 1986).

Besides these references to previous studies of degradation rate, the half-life used in some representative multimedia models can also provide helpful information. The half-life and degradation rate of benzene used in MEPAS, MMSOILS is far different from the ones given by Hutchins et al. (1991a, 1991b), Howard (1990) and Howard et al. (1991). The environmental half-lives of benzene in air, water, and soil are all  $6.9E07$  days

in MEPAS, meaning that the first order decay rates are around 0 in those media. In MMOSILS, the first order decay rate of 0 is used for waste management union, unsaturated zone, groundwater, and air medium.

In addition to theoretical analysis, the monitoring data of TRNL (2002) indicate that the consideration for selecting the degradation rates of BTE is appropriate. Although the Nepean Landfill has been closed for 20 years, there are still BTE concentrations that are being observed in the groundwater very close to the Nepean landfill site right because of a low degradation rate; otherwise, these organic compounds would deplete within a short time.

Judging from the discussion above, it is understood that it is very difficult to estimate a half-life accurately in a landfill chamber, in deep soil, and in groundwater without detailed information about the temperature, the compound composition in the soil, the pollutant concentration in the media, etc. In accordance with the observed BET concentrations in TRNL (2002), it can be estimated that BET have a very low rate of biodegradation in such media.

#### Organic carbon partition coefficient

The organic carbon partition coefficients,  $K_{oc}$ , of BTE can be found in the literature, including HSDB (2005), and Howard (1990) etc. Those coefficients may vary under different conditions such as temperature, pressure. Also, the empirical method can be used to evaluate the value of  $K_{oc}$ . Lyman et al. (1990) and Fetter (1993) give a few available methods for the estimation of  $K_{oc}$  expressed in log-log form:

$$\log K_{oc} = a_1 \log(S_o, K_{ow}, \text{or } BCF) + b_1 \quad [5.1]$$

where  $a_1$  and  $b_1$  are constants;  $S_o$  is the chemical- water solubility (mol/litre);  $K_{ow}$  is the octanol-water partition coefficient (dimensionless); BCF is the bioconcentration factor for aquatic life (dimensionless).

In this study, Lyman's (1990) and Fetter's (1993) estimation methods are used. The detailed formulas and calculations are presented below.

The  $K_{oc}$  of benzene is estimated by using the one of the methods given in Lyman et al. (1990):

$$\log K_{oc} = 0.544 \log(K_{ow}) + 1.377 \quad [5.2]$$

The  $K_{oc}$  of toluene is calculated by using the equation in Fetter (1993):

$$\log K_{oc} = 1.00 \log(K_{ow}) - 0.317 \quad [5.3]$$

The  $K_{oc}$  of ethylbenzene is estimated using the equation in Fetter (1993):

$$\log K_{om} = 0.52 \log(K_{ow}) + 0.62 \quad [5.4]$$

and

$$K_{oc} = 1.724 * K_{om} \quad [5.5]$$

All the employed values of  $K_{ow}$  and the calculated  $K_{oc}$  are given in the Table 5.2.

Table 5.2 Estimation of the organic carbon partition coefficient ( $K_{oc}$ )

Chemical	$\log K_{ow}$	Reference	$K_{oc}$ (m <sup>3</sup> /kg)	Reference
Benzene	2.13	HSDB 2005	0.347	Lyman et al. 1990
Ethylbenzene	3.13	Yalkowsky and Valvani 1976	0.305	Fetter 1993
Toluene	2.72	Howard 1990	0.25	Fetter 1993

### Henry's law constant

Henry's law constant can be found in much of the literature, including HSDB (2005), Howard (1990) etc. This constant varies with environmental conditions. Generally, it is expressed in the unit of atm-m<sup>3</sup>/mol in the reference. But it is used in the form

dimensionless in the present study. The relationship between the two forms of Henry's law constant is described as follows:

$$\text{Henry's law constant (dimensionless)} = \frac{\text{Henry's law constant (atm m}^3 / \text{mol)}}{8.2057 \times 10^{-5} (\text{m}^3 \text{ atm} / \text{mol K}) \times 298(\text{K})} \quad [5.6]$$

The Henry's law constants of BTE for the present study are expressed in Table 5.3.

Table 5.3 Estimation of Henry's law constant

	Henry's law constant (atm-m <sup>3</sup> /mol)	Henry's law constant (dimensionless)	Reference
Ethylbenzene	7.9E-3	0.323	Mackey and Shiu 1981
Toluene	5.94E-3	0.243	Howard 1990
Benzene	5.5E-3	0.225	Mackay and Leinonen 1975

#### Organic carbon fraction

An organic carbon fraction specifies the fraction of the organic carbon in the contaminated material that is used to estimate the chemical distribution coefficient between the solid and liquid phase (Solhotra et al. 1995, Lin and Hildemann 1995, Jury et al. 1990). Lin and Hildemann (1995) assumed it as 0.0125 in their study under a typical condition. Labieniec et al. (1996a) estimated it as 0.0075 in their case. Based on the data provided by TRNL (2002) and the analysis above, the organic carbon fraction in the present study is 0.0105.

#### **5.2.3.2 Data of the landfill**

##### Gaseous diffusion coefficient

The gaseous diffusion coefficient of the contaminant in air is estimated using the method of Wilkes and Lee, a method recommended by Lyman et al. (1990):

$$D_g^a = \frac{B' T^{3/2} \sqrt{M_r}}{P \sigma_{AB}^2 \Omega} \cdot 8.64 \text{ (m}^2/\text{d)} \quad [5.7]$$

where,  $M_r = \frac{(M_A + M_B)}{M_A M_B}$ ;  $M_A$  is the molecular weight of air;  $M_B$  is the molecular weight of the compound of interest;  $P$  is the pressure (atm);  $T$  is temperature (K), and the parameters  $B'$ ,  $\sigma_{AB}$ , and  $\Omega$  is defined as:

$$B' = 0.00217 - 0.0005 \sqrt{\frac{1}{M_A} + \frac{1}{M_B}} \quad [5.8]$$

$$\sigma_{AB} = \frac{3.711 + 1.18(V_B')^{1/3}}{2} \quad [5.9]$$

$V_B'$  is the LeBas molar volume of the compound of concern in  $\text{cm}^3/\text{mole}$  which can be determined using Table 17-5 in Lyman et al. (1990).

$$\Omega = \frac{a}{(T^*)^b} + \frac{c}{\exp(T^* d)} + \frac{e}{\exp(T^* f)} + \frac{g}{\exp(T^* h)} \quad [5.10]$$

where the values of a-h are as follows:  $a=1.06036$ ;  $b=0.15610$ ;  $c=0.19300$ ;  $d=0.47635$ ;  $e=1.03587$ ;  $f=1.52996$ ;  $g=1.76474$ ;  $h=3.89411$ ;

and

$$T^* = \frac{T}{\sqrt{78.6 * 1.15 T_b}} \quad [5.11]$$

in which,  $T_b$  is the boiling point of the compound (K).

By using Equations [5.7] to [5.11], the BET's gaseous molecular diffusion coefficients are calculated and listed in Table 5.4

Table 5.4 Estimation of gaseous molecular diffusion coefficient ( $D_g^a$ )

Component	Molecular Weight Mw	Boiling point (°C)	LeBas molar volume $V_B'$ (cm <sup>3</sup> /mole)	$D_g^a$ (m <sup>2</sup> /d)	Reference
Benzene	78	80.1	111	0.789	Lyman et al. 1990 and HSDB 2005
Ethylbenzene	106.2	136.2	155.4	0.644	Lyman et al. 1990 and HSDB 2005
Toluene	92.13	110.6	133.2	0.706	Lyman et al. 1990 and HSDB 2005

#### Liquid Molecular diffusion coefficient

The Hayduk and Laudie method presented in Lyman et al. (1990) is employed to calculate the diffusion coefficient of the chemical of interest in water.

$$D_l^w = \frac{13.26 \times 10^{-5}}{\eta_w^{1.14} V_B'^{0.589}} \cdot 8.64 \text{ (m}^2/\text{d)} \quad [5.12]$$

where  $\eta_w$  is the viscosity of water in centipoise at the temperature of concern.

According to the methods above, the liquid molecular diffusion coefficients for BET are calculated and shown in Table 5.5.

Table 5.5 Estimation of liquid molecular diffusion coefficient ( $D_l^w$ )

Component	$\eta$	$V_B'$	$D_l^w$	Reference
Benzene	0.8904	111	8.16272E-05	Lyman et al. 1990
Ethylbenzene	0.8904	155.4	6.69523E-05	Lyman et al. 1990
Toluene	0.8904	133.2	7.33157E-05	Lyman et al. 1990

#### Gaseous velocity

This term is neglected in the model developed by Jury et al. (1983, 1990). However, the previous studies indicate that although it is hard to measure or estimate, it should not be neglected in the mathematical model due to its high values and its “sweep out” impacts



on the gas emission rate. Rickabaugh (1990) measured the gas velocity ranging from 0.1457 to 0.7899 cm/d through the experiment landfill. The study of Lin and Hildemann (1995) indicated that the gaseous velocity in the landfill chamber could be equal to, less than or even greater than the leachate velocity. Furthermore, the study of Thibodeaux et al. (1981) indicates that the velocity of biogas could increase the emission rate by a factor of 7 as the gas velocity within the landfill increases from zero to  $1.63\text{E-}3$  cm/s.

Therefore, it is reasonable to account for the gaseous velocity in the landfill modelling. It is estimated as  $2.6\text{E-}3$  m/d in this model validation based on field data and the analysis above.

#### Leachate velocity

Leachate velocity is approximately equal to the seepage velocity in the unsaturated zone. The detailed calculation is given in the next subsection.

### **5.2.3.3 Soil condition**

#### Dispersivity and diffusion coefficient

According to the tabulated data of longitudinal dispersivity versus scale given by Wilson et al. (1990), with the depth of contaminated site (from the centre of the landfill to the water table), the longitudinal dispersivity falls in a big range from 1.37 m to 98 m. It is difficult to estimate a reasonable value based on such a range. Thus, the longitudinal diffusion coefficient, the value of  $0.24\text{ m}^2/\text{d}$ , is estimated based on the environmental conditions investigated by TRNL (1995, 2002).

The typical relationship (Solhotra et al. 1990) is used to estimate the transverse diffusion coefficient, which is expressed by:

$$\text{transverse diffusion coefficient} = 0.1 * \text{longitudinal diffusion coefficient} \quad [5.13]$$

It is  $0.024 \text{ m}^2/\text{d}$  in this case

#### Seepage velocity

Darcy's law is used to estimate the seepage velocity in the unsaturated zone.

$$V = -\frac{K}{\phi_{un}} \frac{dh}{dx} \quad [5.14]$$

where  $K$  is the hydraulic conductivity (m/d);  $\frac{dh}{dx}$  is the hydraulic gradient;  $\phi_{un}$  is the effective porosity in the unsaturated zone (%).

According to TRNL (2002), the vertical hydraulic conductivity beneath the Trail Road Landfill is  $1.4\text{E-}6\text{m/s}$ , and the average hydraulic gradient under the site is 0.008 with a porosity of 0.3. Thus the average velocity of the fluid is calculated as:

$$V = -\frac{1.4 \times 10^{-6}}{0.3} \times -0.008 \times 3600 \times 24 = 3.23 \times 10^{-3} \text{ m/d}$$

#### **5.2.3.4 Groundwater condition**

##### Dispersivity

Equations for approximating the longitudinal dispersivity in groundwater, which are generally based on field observations, use a characteristic length,  $L_c$ . Below are the equations used in the dispersivity calculation (Xu and Eckstein 1995, Arya 1986).

$$\alpha_L = 0.83(\log_{10} L_c)^{2.414} \quad [5.15]$$

$$\alpha_L = 0.177(L_c)^{0.728} \quad [5.16]$$

where  $\alpha_L$  is the longitude dispersivity in the groundwater (m);  $L_c$  is the characteristic length (m).

The receptor concerned in this case study is 50 m away from the landfill site; thus  $\alpha_L$  is around 3 m calculated by Equations [5.15] and [5.16].

An empirical relationship is used to calculate the transverse dispersivity and the vertical dispersivity (Solhotra et al. 1990):

$$\text{Transverse dispersivity (m)} = \text{longitudinal dispersivity}/3; \quad [5.17]$$

$$\text{Vertical dispersivity (m)} = 0.56 * \text{longitudinal dispersivity}. \quad [5.18]$$

$$\text{Thus } \alpha_T = 1m \quad \alpha_V = 1.68m$$

### Seepage velocity

According to the TNRL (2002), the average horizontal hydraulic gradient under the Trail landfill site is approximated as 0.05, and the average horizontal hydraulic conductivity is estimated as  $4E-5$  m/s with a porosity of 0.3. Thus, by using Equation [5.14] and substituting the  $\phi_{un}$  with  $\phi_s$  the seepage velocity in the groundwater is calculated as:

$$V = -\frac{4 \times 10^{-5}}{0.3} \times -0.05 \times 3600 \times 24 = 0.576m/d$$

#### 5.2.4 Background Contaminant Concentration

The operation period for Stage 1 of Trail Road Landfill is 1980-1986 and 1986-1991 for Stage 2 (TNRL 2002). Accordingly, the average age of Stages 1 and 2 is 6 years. Stage 2 was closed in 1991; thus we consider 1991 as the starting point of the evaluation period.

From the borehole logs schematic in TNRL (2002), the average depth of Stages 1 and 2 of the Trail Road Landfill is estimated as 14 meters. Thus the volume of Stages 1 and 2 of the landfill site is  $5.74\text{E}6 \text{ m}^3$ .

Given that the average annual amount of landfilled waste is approximately 287,000 tonnes during the 12 years of operation, the total amount of landfilled waste in Stages 1 and 2 is  $3.44\text{E}6$  tonnes.

Consequently, the density of the waste in Stages 1 and 2 is  $600\text{kg/m}^3$ .

According to TNRL (1995), the density of the landfilled waste is  $724 \text{ kg/m}^3$  in 1993,  $581 \text{ kg/m}^3$  in 1994, and  $630 \text{ kg/m}^3$  in 1995. Thus, the calculated density for the validation is in this range, confirming that the data related to the volume of the landfill chamber and the weight of the landfilled waste are reasonable.

From the data obtained by theoretical methods and experiments, it can be concluded that the actual gas generation potential of landfills varies in the range of  $100\text{--}250\text{m}^3/\text{tonne-wet-refuse}$  (Nastev 1998). The average value of  $175 \text{ m}^3/\text{tonne}$  is used for the Trail Road Landfill based on field observation. Therefore, the initial gas generation potential of Stages 1 and 2 is  $6.03\text{E}8 \text{ m}^3$ .

There are two approaches for the mathematical presentation of the gas generation potential: the zero-order kinetic model and the first-order kinetic model. The

first-order kinetic gas generation potential is the most common representation of gas generation potential. Many scientists agree with that. It is thus employed for the Trail Road Landfill.

$$\frac{dS}{dt} = -kS \quad [5.19]$$

where  $S$  is the gas generation potential ( $\text{m}^3$ );  $k$  is a fitting parameter.

If the initial gas generation potential is  $S_0$ , then it can be obtained as:

$$S = S_0 \exp(-kt) \quad [5.20]$$

Substituting  $S_0 = 6.03\text{E}8 \text{ m}^3$ , and the  $S=0 \text{ m}^3$  at  $t= 50 \text{ yrs}$  into Equation [5.20],  $k=2.5 \text{ (1/yr)}$  can be calculated. Equation [5.20] becomes

$$S = S_0 \exp(-2.5t) \quad [5.21]$$

Taking  $t=6 \text{ yr}$  into Equation [5.21], the remaining gas generation potential in landfill for Stages 1 and 2 is  $4\text{E}-7S_0$ .

The typical constituents and compounds suggested by Tchobanoglous et al. (1993) are referred to in order to estimate the concentration of trace compounds in the Trail landfill.

Table 5.6 Typical constituents of landfill gas

Component	Percent (dry volume basis)
Methane	40-60%
Carbon Dioxide	40-60%
Nitrogen	2-5%
Oxygen	0.1-1.0%
Ammonia	0.1-1.0%
Sulfides, disulfides, mercaptans, etc.	0-0.2%
Hydrogen	0-0.2%
Carbon Monoxide	0-0.2%
Trace Constituents	0.01-0.6%

Source: (Tchobanoglous et al. 1993)

Table 5.7 Typical distribution of the trace constituent

Component	Mean Concentration (pbV, parts per billion by volume)	Percent (trace constituent basis)
Toluene	34,907	34.3%
Dichloromethane	25,694	25.2%
Ethylbenzene	7,334	7.2%
Acetone	6,838	6.7%
Vinyl Acetate	5,663	5.6%
Tetrachloroethylene	5,244	5.1%
Vinyl Chloride	3,508	3.4%
Methyl Ethyl Ketone	3,092	3.0%
Xylenes	2,651	2.6%
1,1-Dichloroethane	2,801	2.7%
Trichloroethylene	2,079	2.0%
Benzene	2,057	2.0%

Source: (Tchobanoglous et al. 1993)

The trace constituent of Trail landfill gas is 0.5%. It is estimated that benzene accounts for 0.02\*0.5% of the total landfill gas; ethylbenzene accounts for 0.10\*0.5%; and toluene accounts for 0.33\*0.5%. With the density of three components is shown as below (HSDB 2005 and Spectrum data base): the density of benzene is 879 kg/m<sup>3</sup>; that of ethylbenzene is 867 kg/m<sup>3</sup>; and that of toluene is 866kg/m<sup>3</sup>. The background benzene concentration in the landfill site in 1991 is calculated as:

$$C_{L0} = \frac{6.03 \times 10^8 (m^3) \times 4 \times 10^{-7} \times 0.5\% \times 2\% \times 879 (kg / m^3) \times 10^9 (\mu g / kg)}{3.44 \times 10^6 (tonnes) \times 10^6 (g / tonne)} = 0.0062 \mu g / g$$

Similarly, the toluene is 0.1  $\mu g / g$  ; and the ethylbenzene is 0.032  $\mu g / g$  .

### 5.2.5 A Summary of the Input Parameters and Data

In summary, the input parameters related to chemical properties are exhibited in Table 5.8, and other parameters associated with environmental conations and the physical properties of the site are summarized in Table 5.9.

Table 5.8 Input parameters related to chemical properties

Property	Symbol	Benzene	Ethylbenzene	Toluene
The gaseous diffusion coefficient in air	$D_g^a$ (m <sup>2</sup> /d)	0.756	0.635	0.706
The liquid diffusion coefficient in water	$D_l^w$ (m <sup>2</sup> /d)	$8.81 \times 10^{-5}$	$6.7 \times 10^{-5}$	$7.33 \times 10^{-5}$
Organic carbon partition coefficient	$K_{oc}$ (m <sup>3</sup> /kg)	0.347	0.305	0.25
Henry's law constant, dimensionless	$K_H$	0.225	0.322	0.25
Half-life in landfill	$\tau$ (d)	-	-	-

Table 5.9 Physical properties and environmental conditions in the landfill site

Parameters in landfill module					
Length orthogonal to groundwater flow.	$A_y$ (m)	500	Length parallel to groundwater flow	$A_x$ (m)	800
The volumetric air content of the soil	$\alpha$	0.2	Volumetric water content at field capacity	$\theta$	0.3
Organic carbon fraction	$f_{oc}$	0.0105	Bulk density	$\rho_b$ (kg/m <sup>3</sup> )	600
The thickness of landfill cover	$d$ (m)	1	Landfill depth	$L$ (m)	14
Gaseous velocity	$v_g$ (m/d)	2.6E-3	Leachate velocity	$v_L$ (m/d)	3.23E-3
Parameters in unsaturated zone module			Parameters in saturated zone module		
Coefficient of longitudinal dispersion	$D_L$ (m <sup>2</sup> /d)	0.24	Darcy velocity	$V_d$ (m/d)	0.576
Coefficient of transverse dispersion	$D_T$ (m <sup>2</sup> /d)	0.024	Bulk density	$\rho_{sat}$	1350
Average velocity of fluid	$v$ (m/d)	3.23E-3	Porosity	$\phi_s$	0.3
Porosity	$\phi_{un}$	0.365	Organic carbon fraction	$f_{ocsat}$	0.0105
Bulk density of unsaturated zone	$\rho_{unsat}$ (kg/m <sup>3</sup> )	1350	Half-life	$\tau_{sat}$ (d)	-
Half-life in unsaturated zone	$\tau_{unsat}$ (d)	-	Dispersion coefficient in x direction	$D_x$ (m <sup>2</sup> /d)	1.728
Water table depth	$z_{wt}$ (m)	10	Dispersion coefficient in y direction	$D_y$ (m <sup>2</sup> /d)	.576
Organic carbon fraction	$f_{ocun}$	0.0105	Dispersion coefficient in z direction	$D_z$ (m <sup>2</sup> /d)	0.968
Parameters in air module.					
Annual wind speed frequency	$f(\phi)$	0.13	Wind speed	$w$ (m/s)	4.3

### 5.2.6 Field Modelling and Validation

Modelling simulation is applied to the Trail Road Landfill at monitoring stations M32 and M90 whose locations are shown in Figure 5.4. All the input parameters are entered into the multimedia modelling system, and they will then be automatically utilized by the modules in accordance with the internal order of the model execution.

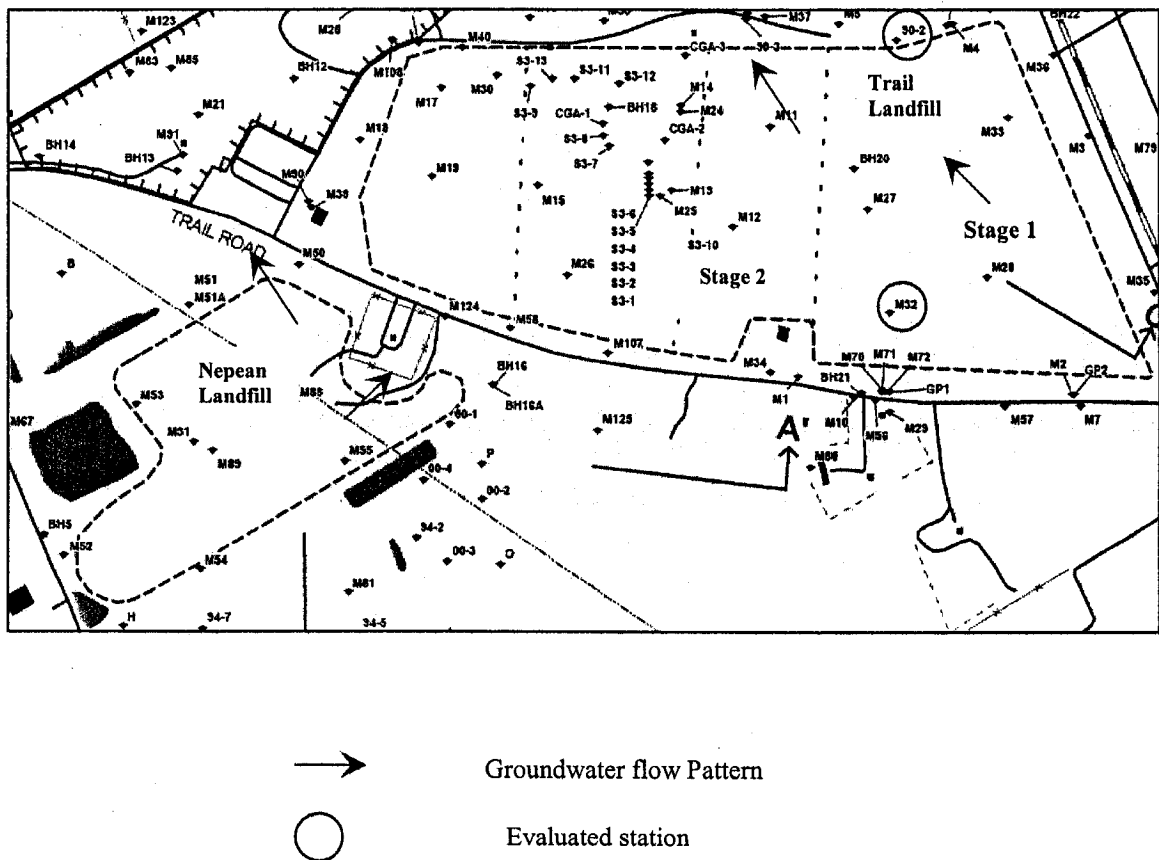


Figure 5.4 Location of monitoring station (TRNL 2002)



### 5.2.7 Simulation Results and Comparison With Observed Data

Detailed modelling results for the contaminants of ethylbenzene in Trail Road landfill are given in Table 5.10.

Table 5.10 Comparison of modelling results and observed ethylbenzene concentrations at the monitoring well M32

Year	Modelled concentration at M32 (mg/m <sup>3</sup> )	Observed concentration at M32 (mg/m <sup>3</sup> )
1999	29.5	31.1
2000	28.8	30.5
2001	28.1	1.8
2002	27.2	15.6

The simulated concentration of ethylbenzene in the groundwater at  $x=0$  gradually decreased from 29.5 mg/m<sup>3</sup> in 1999 to 27.2 mg/m<sup>3</sup> in 2002 due to spreading via dispersion in soil and groundwater and a little loss via volatilisation from the landfill cover. The decrease in the concentration is mainly achieved thanks to the annual infiltration rate through the landfill cover and thanks to chemical properties. Ethylbenzene is not a highly volatile compound, implying that volatilisation is not a significant loss pathway for this compound; on the other hand, it does migrate downward with infiltration and does enter into the groundwater.

The simulated concentrations are in an agreement with the observed concentrations in 1999 and 2000. There are some differences for the years 2001 and 2002. The reason may lie in that the precipitation rate in 2001 was smaller than that in 1999 and 2000; more contaminant thus remained in the soil in that year than in the

previous two years. After the infiltration rate rose again, the retarded contaminant was solved, and then along with the newly arriving pollutant leachate from the landfill was carried to the groundwater. As a result, the concentration at this location went up from 1.8 mg/m<sup>3</sup> to 15.6 mg/m<sup>3</sup>. The discrepancies stem from (1) a significant change in the precipitation in 2001; (2) sampling and chemical analysis errors; and (3) complexities in the site conditions.

Table 5.11 Comparison of modelling results and observed toluene concentrations at the monitoring well M32

Year	Modelled concentration at M32 (mg/m <sup>3</sup> )	Observed concentration at M32 (mg/m <sup>3</sup> )
1999	106	92
2000	103.4	108
2001	100.3	21.6
2002	96.8	54.5

The simulated toluene concentration in the groundwater at M32 is shown in Table 5.11. It decreased quickly from 106 mg/m<sup>3</sup> in 1999 to 96.8 mg/m<sup>3</sup> in 2002. The simulated results are almost as same as the observed ones in 1999 and 2000, but they are significantly different from the observed values in 2001 and 2002. The reason for this phenomenon is the same as that for the ethylbenzene.

Table 5.12 Comparison of modelling results and observed benzene concentrations at the monitoring well M90

Year	Modelled concentration at M90 (mg/m <sup>3</sup> )	Observed concentration at M90 (mg/m <sup>3</sup> )
1999	6.1	6.3
2000	6.0	6.2
2001	5.8	9.8
2002	5.7	3.5

The concentration of benzene at M92 decreased slowly from 6.1 mg/m<sup>3</sup> in 1999 to 5.7 mg/m<sup>3</sup> in 2002. Dispersion explains the main pathway loss, while the volatilization of the benzene was not due to its modest Henry's law constant and the protection of the landfill cover.

The modelling results are very close to the observed concentration at the monitoring station in 1999 and 2000. But the monitoring data oscillates in the following years and shoes a trend that is different from those of the toluene and ethylbenzene at station M32. That may be due to the diverse geological conditions under the landfill site. A greater amount of pollutant was carried to this point but could not be swept away due to the low groundwater flow and the retardation of the soil.

Real-world environmental system modelling is a difficult task. In this present study, a field-scale model validation has been conducted. Reasonable results have been obtained. The modelling outputs indicate that the developed EMMS enables us to investigate large-scale site contamination with multimedia characteristics.

On the other hand, the large number of various uncertainties involved significantly limits the capacity of the modelling tool. In order to mitigate this limitation,

the developed fuzzy-set enhanced EMMS is utilized to conduct an uncertainty analysis and an uncertainty quantification for this field validation in the next section.

## 5.3 Uncertainty Analysis

The fuzzy set methodology introduced in chapter 3 is applied in this section to deal with the uncertainties associated with both individual and interacted parameter values, and the description of the uncertainties is given in the form of level of confidence range.

### 5.3.1 Quantification of Uncertainties Using the Fuzzy-Set Approach

The uncertainties of the parameters in the landfill module along with the unsaturated zone module, including those associated with chemical properties, landfill setting, and environmental conditions, are first reflected in Equation [3.42]. Therefore, the application of the fuzzy method to this module as a starting point reduces the uncertainties of the multimedia modelling system to a large extent.

In details, Equations [3.40] and [3.43] can be converted to the form of:

$$J_l(z, t) = m_0 TR_u \quad [5.22]$$

in which  $m_0$  is rate of release of the solute from the landfill bottom at the starting point of the evaluation period (g/d).  $TR_u$  is the remaining part of the equation, named the transfer function in this study, and its combined unit is  $m^{-2}$ .

$m_0$  is derived from the landfill module, reflecting the uncertainty caused by the estimations of the chemical properties, of the physical and dynamical properties and of

the layout of the landfill site. It is reasonable to consider it as a fuzzy number.  $TR_u$  is deduced from the unsaturated zone module. The uncertainty caused by the dispersivity and the infiltration rate, both of which occupy the central role to assure the accuracy of model outputs, can be dealt with through fuzzifying  $TR_u$ .

After the multiplication operation is applied to Equation [5.22], fuzzy set outputs can be obtained, and then in turn as fuzzy inputs entry into the saturated zone module. In the saturated zone module, similar operations are implemented. Eventually, fuzzy-set enhanced predictions are produced by the EMMS. The detailed steps are given below:

1) Equations [3.40] and [3.43] are transformed to the form described by Equation [5.22]

2) The operations of fuzzy numbers are applied to Equation [3.42], sequentially; the integration of  $J_{un}$  in terms of  $t$ ,  $M_{un}$ , can be treated as  $\sum_0^t J_{un} \Delta t$ . Equation [3.44] can then be solved by applying an addition operation.

3) In Equation [3.46], all the parameters except  $M_{un}$  are treated as crisp numbers due to their negligible influence on the model results. Similarly, Operation 3 of fuzzy numbers is applied to Equation [3.46]. Then, a fuzzy output,  $C_{s0}$ , can be calculated and input into the saturated zone module.

Similarly, Equation [3.48] can be transformed to:

$$C_s(x, y, z, t) = C_{s0} TR_s \quad [5.23]$$

Similar to  $TR_u$ ,  $TR_s$  is a systematic dimensionless coefficient reflecting the ambiguous information associated with the dispersivity and seepage velocity in the

saturated zone, and can be considered as a fuzzy number to deal with the uncertainties of the model outputs resulting from an interaction between important parameters.

Following the application of the multiplication of fuzzy numbers to Equations [5.22] and [5.23], the fuzzy outputs of the multimedia model are computed. Comparing the observed data with the fuzzy outputs, it is easy to see whether the observed data fall in the predicted uncertainty range.

The detailed applications of the fuzzy set to EMMS are summarized in Figure 5.5.

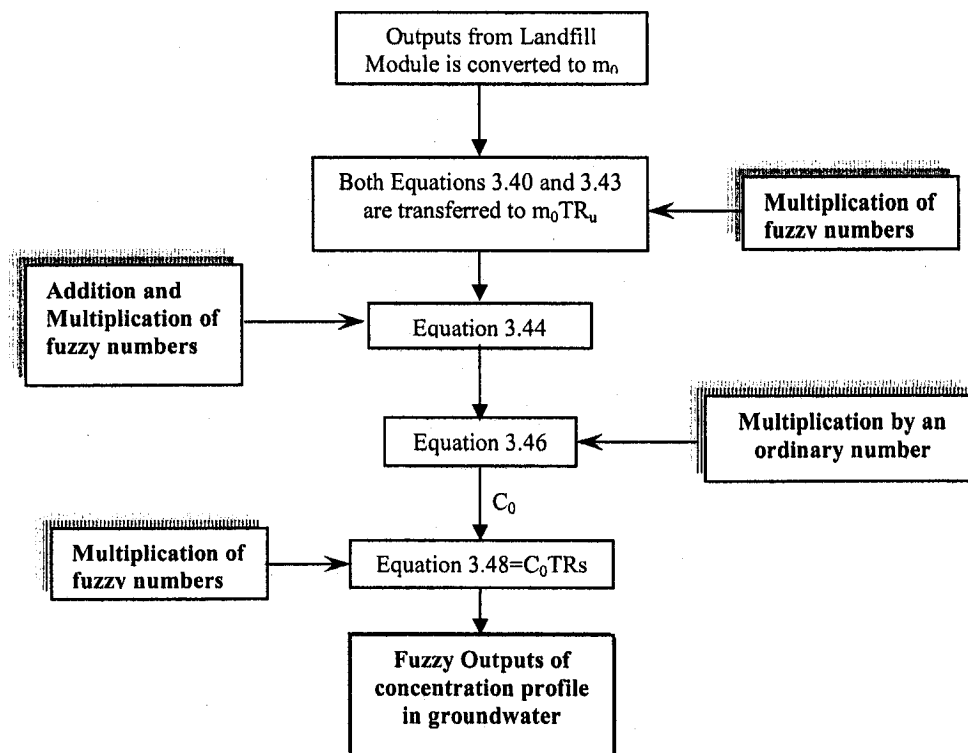


Figure 5.5 Fuzzy analyses to quantify uncertainties associated with leachate transport process

### 5.3.2 Uncertainty Analysis on the Modelling of Trail Road Landfill

Following the scheme presented in Figure 5.5, uncertainty analysis methodology is applied to EMMS to produce fuzzy outputs. Detailed modelling results are exhibited in Tables 5.13 to 5.15. Using Equation [3.55], we can calculate the membership of observed data falling into the interval confidence of the fuzzy output.

Table 5.13 The results of uncertainty analysis for ethylbenzene

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M32 (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )	Observed concentration (mg/m <sup>3</sup> )	The possibility degree
1999	16.2	29.5	42.8	31.1	0.88
2000	15.9	28.8	41.8	30.5	0.87
2001	15.4	28.1	40.7	1.8	-
2002	15.0	27.2	39.4	15.6	0.05

The simulated mean concentration of the ethylbenzene beneath the Trail Road Landfill in 1999 was 29.5 mg/m<sup>3</sup> and gradually decreased to 27.2 mg/m<sup>3</sup> in 2002; the low bond concentration decreased correspondingly from 16.2 mg/m<sup>3</sup> to 15.0 mg/m<sup>3</sup>; and the high bond concentrations show the same trend from 42.8 mg/m<sup>3</sup> to 39.4 mg/m<sup>3</sup>.

Compared with the observed concentration in the groundwater beneath the Trail Road Landfill, the first two calculated mean concentrations agree significantly with the monitoring data, which get 0.88 and 0.87 of grade of membership in the fuzzy set outputs. The monitoring data in 2000 is out of the range of the fuzzy set output, since it sharply decreased to 1.8 mg/m<sup>3</sup> and then rose to 15.6 mg/m<sup>3</sup>. The reason for these

abnormal decreases may result from variations in the flow of the groundwater, from the infiltration rate in those years, and also from the uncertainty of gas generation in the landfill chamber.

Table 5.14 The results of uncertainty analysis for toluene

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M32 (mg/m <sup>3</sup> )	High bound of concentration (mg/m <sup>3</sup> )	Observed concentration at M32 (mg/m <sup>3</sup> )	The possibility degree
1999	58.3	106	153.7	92	0.71
2000	56.9	103.4	149.9	103	0.90
2001	55.2	100.3	145.4	21.6	-
2002	53.2	96.8	140.3	54.5	0.03

The simulated concentration of toluene is 106 mg/m<sup>3</sup> in 1999 and then decreases to 96.8 mg/m<sup>3</sup> in 2002; the low bound varies from 58.3 mg/m<sup>3</sup> to 53.2 mg/m<sup>3</sup>; and the high bound similarly shows a decreasing pattern from 153.7 mg/m<sup>3</sup> to 140.3 mg/m<sup>3</sup>.

According to the comparisons for toluene, the observed data in 2001 is slightly out of the predicted range, while all the other three monitoring concentrations nicely fall into the simulated ranges, showing high degrees of membership. It indicates that the modelling results are compatible with the monitoring results. The fluctuation of the concentration in 2001 may be caused by weather conditions and the hydraulic environment of that year as discussed before.



Table 5.15 The results of uncertainty analysis for benzene

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M90 (mg/m <sup>3</sup> )	High bound of concentration (mg/m <sup>3</sup> )	Observed concentration at M90 (mg/m <sup>3</sup> )	The possibility degree
1999	3.4	6.1	8.8	6.3	0.93
2000	3.3	6.0	8.7	6.2	0.93
2001	3.2	5.8	8.5	9.8	-
2002	3.1	5.7	8.3	3.6	0.15

The range of benzene in the groundwater 50 meters away from the Trail Road Landfill site is from 6.1 mg/m<sup>3</sup> to 5.7 mg/m<sup>3</sup>; the low bond was 3.4 mg/m<sup>3</sup> in 1999 and 3.1 mg/m<sup>3</sup> in 2002; and the high bond was from 8.8 mg/m<sup>3</sup> to 8.3 mg/m<sup>3</sup> during these 4 years.

Although the observed benzene concentration at M92 does not fall into the computed set, all others show high agreement with the model results. Especially, the simulated concentrations for 1999 and 2000 are almost as same as those observed on site. The observed data of 2001 is out of the decreasing trend probably due to the meteorological and hydraulic condition of that year as discussed before.

## 5.4 Summary

EMMS is applied to the Trail Road Sanitary Landfill to validate the model performance and the suitability of the application. The observed contaminant concentrations from the groundwater in the vicinity of the Trail Road Landfill are used to examine the simulated pathway from the landfill to the groundwater.

The results from the comparison study are satisfactory. Most of the modelling yields closely match the monitoring data collected from sites. Besides an innovative design of an EMMS, a thorough consideration of model parameters and data collection/estimation, and a systematic execution of EMMS, a fuzzy-set approach has been applied to quantify uncertainties associated with the field-scale study to further validate the developed environmental multimedia modelling system in this section.

## **Chapter 6**

# **Development of a User-friendly Engineering Interface and Field Application**

The developed EMMS is tested and validated in Chapters 4 and 5. Aiming to make the modelling system user-friendly, a user interface is designed for the EMMS in this chapter. The developed EMMS thus can become a user-friendly fuzzy-set enhanced EMMS. To extend the model testing, validation and field-scale study, the user-friendly system is applied to the Trail Road Landfill to assess future risks associated with the ambient air, soil and groundwater.

### **6.1 Design of the Graphic User Interface (GUI) Supported EMMS**

Besides the efforts to develop a new EMMS in terms of functionality and engineering applicability, the system is also designed for better computational efficiency and for user-friendliness, compared to other environmental multimedia models. Figure 6.1 presents an

overview of the system design for EMMS. The system includes the following components: fate and transport modules, a system graphic user interface (GUI), a system input processing unit, and a system output processing function. The GUI is a tool to help users use the model and process data. More details are given in the subsequent sections.

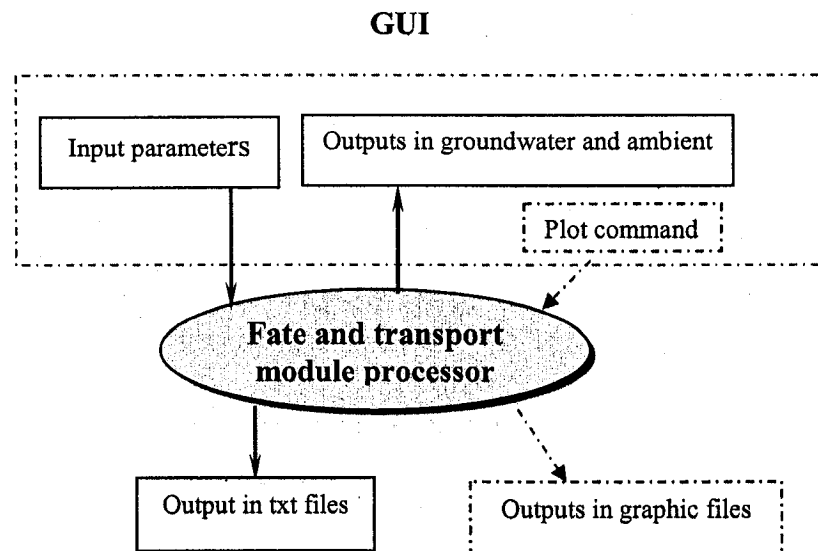


Figure 6.1 The schematic of system design

### 6.1.1 Engineering User Interface Design

For easy operation of the modelling system without iterative work, a graphic user interface (GUI) has been designed using Matlab program code. The user interacts with the GUI by communicating requirements into EMMS and manipulating functional modules to obtain the expected results. A set of initial input data is created in the system and is also displayed in the GUI as shown in Figure 6.2. The concentrations in the groundwater and the ambient air in Figure 6.2 are established through appropriate

modules and those inputs. The user can enter the needed input parameters for a special case situation and the corresponding results are automatically presented in the “output window” of the GUI. The graphic results are displayed in a series of figures after pushing the “plot now”.

The screenshot displays the 'multimedia\_interface\_fuzzy' GUI with several input modules and output sections.

**Chemical Property Input Data:**

$K_p$	0.22	$D_{ij}(m^2/s)$	$8.162 \times 10^{-5}$
$E_{ij}$	-	$D_{os}(m^2/s)$	0.7889
$T_{loc}$	0.0105	$K_{oc}(m^3/kg)$	0.347

**Input Data for Soil Module:**

$x(m)$	14	$\alpha(m/s)$	0.00323	$\beta(m/s)$	0.0062
$x(m)$	14	$\alpha(m/s)$	0.0026	$\beta(m/s)$	0.3
$x(m)$	1	$\alpha(m/s)$	600	$\beta(m/s)$	0.2

**Input Data for Distribution:**

$D_{ij}(m^2/s)$	1.728	$distance(m)$	50	$Q_{ij}(m^3/s)$	0.576
$D_{ij}(m^2/s)$	0.576	$Q_{ij}(m^3/s)$	0.576	$Q_{ij}(m^3/s)$	0.3
$D_{ij}(m^2/s)$	0.9677	$R(m)$	10	$Q_{ij}(m^3/s)$	1350

**Other Input:**

$time(s)$	21	$R(m)$	800
$A_z(m)$	500	$Q_{ij}(m^3/s)$	0

**Input Data for Air Module:**

$Q_{ij}(m^3/s)$	4.3
$IS$	0.13

**Groundwater Output:**

$x(m)$	0.01	$x(m)$	0.01
Concentration ( $g/m^3$ )	0.00374588		
Low band of concentration ( $g/m^3$ )	0.00207369		
High band of concentration ( $g/m^3$ )	0.00541807		

**Ambient Air Concentration:**

$x(m)$	500
$Q_{ij}(g/m^3)$	$4.19431e-013$
Low band of concentration ( $g/m^3$ )	$2.93602e-013$
High band of concentration ( $g/m^3$ )	$5.4526e-013$

A 'plot now' button is visible on the right side of the interface.

Figure 6.2. An example of the system interface

### **6.1.2 System Input Processing**

Input data can be drawn up into six groups: chemical property, landfill module data, unsaturated zone data, saturated data, and other inputs. These data relate to the site layout, the environmental conditions, the meteorological conditions, and the chemical properties. The detailed information about the definitions and units of these data has been presented in Table 5.9.

### **6.1.3 System Output Processing**

The simulation results can be output in three ways: [1] pollutant concentrations in the groundwater and the ambient receptors presented in the GUI “output window”; [2] the concentration distribution, the inter-media flux for consecutive years in txt files that are divided into landfill file, unsaturated file, groundwater file, and ambient air quality file; and [3] results in [1] and [2] can be displayed in the form of a Matlab figure in the order of the evaluated year after pushing the “plot now” button on the GUI; for example, the figures for a case study in Subsection 4.2.3 are plotted for one year of an evaluation period (see Figure 4.5 to Figure 4.11).

## **6.2 Field Applications of the User-friendly EMMS**

The developed user-friendly EMMS is applied to the Trail Road Landfill in this section. All the input data are adopted from Section 5.2. For example, the modelling input parameters are the same as those in Table 5.8 and Table 5.9. The evaluation period is

extended to 20 years, that is, until 2011. The evaluation locations in the groundwater are still M32 and M90, while it is assumed that there is a receptor positioned in a southwest direction and 500 meters away from the landfill site boundary.

The simulation results contain the contaminant concentration distribution in the environmental media and the inter-media fluxes, and the concentrations at the receptors in the groundwater and atmosphere for each contaminant: benzene, ethylbenzene, and toluene (BET). However, for the comparison of the variation of the contaminants in the groundwater or in the ambient air, the outputs are discussed as two groups: the outputs for ambient groundwater and air quality.

### **6.2.1 Evolution of Groundwater Contaminant in the Tail Road Landfill Site for 2003-2011**

The modelling results are obtained and displayed in Figures 6.3 to 6.5.

#### **6.2.1.1 Modelling results and analysis**

As illustrated in Figure 6.3, the average concentration of benzene decreases slowly from 5.5 mg/m<sup>3</sup> to 3.9 mg/m<sup>3</sup> under the environmental conditions during the 9 years of the evaluation period. That means the benzene concentration at this monitoring point eventually meets the Ontario Drinking Water Standard (ODWS) at 5 mg/m<sup>3</sup> as the pollutant is diluted by the groundwater. Thus, the risk of exposure to benzene diminishes with the time passing if the environmental conditions show no signs of significant changes.

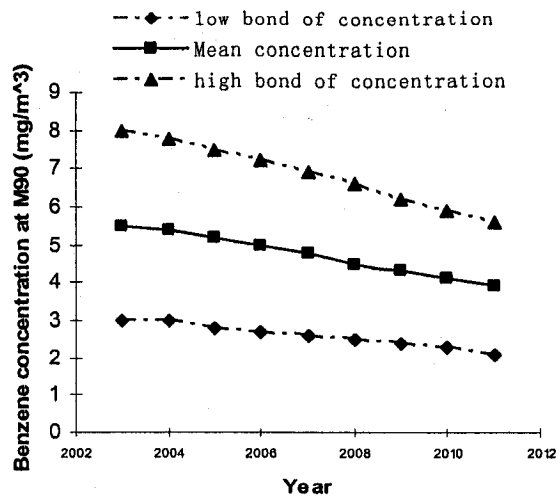


Figure 6.3 Benzene concentrations at the monitoring well M90 for 2003-2011

In Figure 6.4, it can be seen that the mean ethylbenzene concentration varies gradually from 26.3 mg/m<sup>3</sup> to 17.4 mg/m<sup>3</sup>. There is 8.9 mg/m<sup>3</sup> of concentration decrease that occurs due to the groundwater dilution. However, the concentration in 2011 still cannot be lower than the ODWS at 2.4 mg/m<sup>3</sup>.

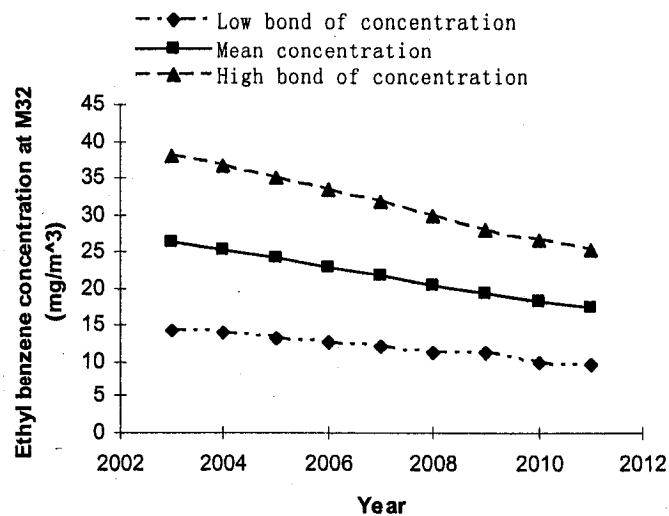


Figure 6.4 Ethylbenzene concentrations at the monitoring well M32 for 2003-2011



Changing a little faster than the previous two pollutants, the mean concentration of toluene decreases slowly from 92.7 mg/m<sup>3</sup> to 56 mg/m<sup>3</sup>. Even 9 years after 2003, the concentration in the groundwater is still high. It exceeds the ODWS at 24 mg/m<sup>3</sup>.

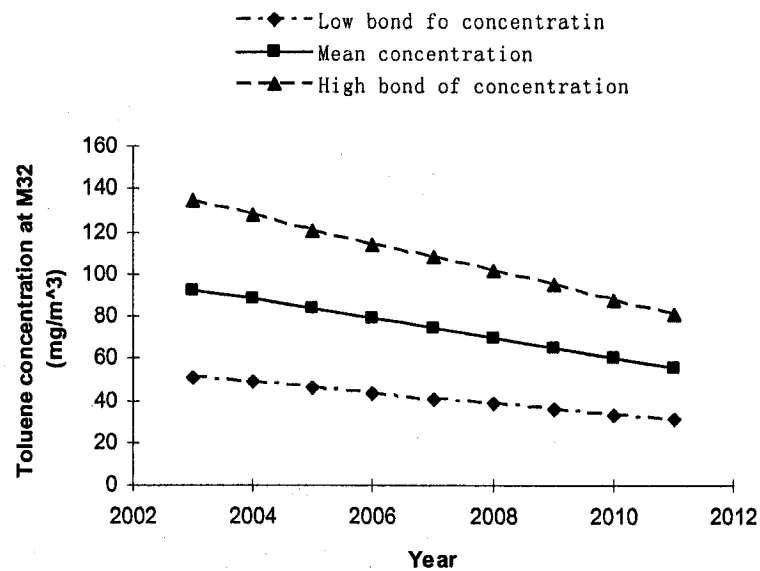


Figure 6.5 Toluene concentrations at the monitoring well M32 for 2003-2011

A comparison of the decreases of contaminants at the evaluation location over a period of 9 years shows that toluene experiences the greatest drop from 92.7 mg/m<sup>3</sup> to 56 mg/m<sup>3</sup>, accounting for 39.6% of the concentration at the start point of the evaluation term. The ethylbenzene decreases from 26.3 mg/m<sup>3</sup> to 17.4 mg/m<sup>3</sup> with a decrease of 33.8% of the concentration during these 9 years. Benzene shows the smallest decrease of concentration, that being 29.1% of the start concentration in 2003. The reasons for the different decreases in these three contaminants are that their organic carbon partition coefficients are different, significantly affecting the chemical partitioning between the liquid and solid phases. They can be arranged in this order: benzene > ethylbenzene > toluene. Therefore,

the greater this order, the more the contaminant is retarded at the solid phase, and the less the amount is carried away by the groundwater flow from this consideration.

#### 6.2.1.2 Predicted uncertainties

Fuzzy outputs of benzene concentrations at M90 are expressed in Table 6.1 and Figure 6.3. The high bound of concentration decreases from 8.0 mg/m<sup>3</sup> to 5.6 mg/m<sup>3</sup>, and the low bound decreases from 3.0 mg/m<sup>3</sup> to 2.1 mg/m<sup>3</sup>. This indicates that the benzene concentration at the end of the evaluation period will fall into the range of 2.1 to 5.6 mg/m<sup>3</sup> if there are fluctuations in the environmental conditions or if there are modelling errors.

Table 6.1 Benzene concentrations at the monitoring well M90 for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M90 (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	3.0	5.5	8.0
2004	3.0	5.4	7.8
2005	2.8	5.2	7.5
2006	2.7	5.0	7.2
2007	2.6	4.8	6.9
2008	2.5	4.5	6.6
2009	2.4	4.3	6.2
2010	2.3	4.1	5.9
2011	2.1	3.9	5.6

The ethylbenzene concentrations are presented in Table 6.2 and Figure 6.4. The high bound of concentration decreases from 38.1 mg/m<sup>3</sup> to 25.2 mg/m<sup>3</sup>, while the low bound decreases from 14.4 mg/m<sup>3</sup> to 9.6 mg/m<sup>3</sup>. At the end point of the evaluation term, the ethylbenzene concentration is between the scope of 9.6 mg/m<sup>3</sup> and 25.2 mg/m<sup>3</sup>, thereby interpreting the fluctuation of the environmental conditions and the discrepancy in the estimates of the parameters. According to the predictions, the low bound of predicted concentration is much greater than the ODWS at 2.4 mg/m<sup>3</sup>. That shows that the risk receptor location under study will still be contaminated after 9 years.

Table 6.2 Ethylbenzene concentrations at the monitoring well M32 for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M32 (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	14.4	26.3	38.1
2004	13.9	25.3	36.6
2005	13.3	24.2	35.1
2006	12.7	23.0	33.4
2007	12.0	21.8	31.7
2008	11.3	20.6	29.9
2009	11.3	19.4	28.1
2010	10.1	18.4	26.6
2011	9.6	17.4	25.2

Table 6.3 and Figure 6.5 exhibit the fuzzy set outputs for the toluene concentrations beneath the Trail Road Landfill site. The high bound of toluene concentration decreases from 134.5 mg/m<sup>3</sup> to 81.2 mg/m<sup>3</sup>, while the low bound decreases from 51 mg/m<sup>3</sup> to 30.8 mg/m<sup>3</sup>. Since the toluene is present in high initial concentration

compared to the other two contaminants, it still ranges between 30.8 mg/m<sup>3</sup> and 81.2 mg/m<sup>3</sup> after 9 years. The low bound of computed concentration is little over the ODWS at 24 mg/m<sup>3</sup>; thus the action should be taken to protect the downstream groundwater.

Table 6.3 Toluene concentrations at the monitoring well M32 for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at M32 (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	51.0	92.7	134.5
2004	48.5	88.2	127.9
2005	45.8	83.3	120.9
2006	43.3	78.7	114.1
2007	40.9	74.4	107.9
2008	38.4	69.9	101.3
2009	35.9	65.3	94.6
2010	33.3	60.6	87.9
2011	30.8	56.0	81.2

## 6.2.2 Evolution of Ambient air Quality in the Trail Road Landfill Site for 2003-2011

By using the same run, the ambient air quality is assessed for the period of 2001 to 2011. The results are given in Figures 6.6 to 6.8.

### 6.2.2.1 Modelling results and analysis

The average benzene concentration in the ambient air rapidly drops from 2.991E-11 mg/m<sup>3</sup> to 8.23E-12 mg/m<sup>3</sup> as presented in Figure 6.6, a significant drop during the 9-year

evolution time. The concentration for each evaluated year at exposure site is far less than the  $8.0\text{E-}2 \text{ mg/m}^3$  of the risk assessment reference concentration (USEPA 1999a, 2002). Thus, it can be concluded that the high-density landfill cover effectively prevents the volatile organic compound from emitting through the landfill surface, and the risk resulting from the inhalation of benzene for the human beings at the exposure site can be neglected.

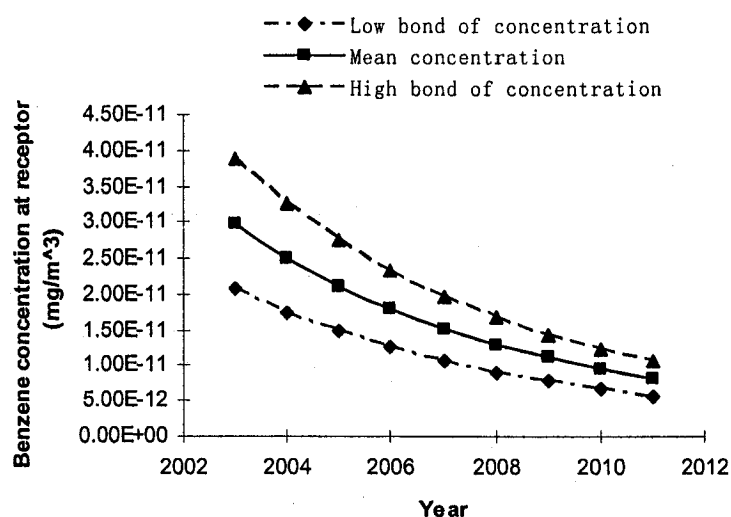


Figure 6.6 Yearly benzene concentration profile at ground surface receptor for 2003-2011

The variance of ethylbenzene concentration in the atmosphere near the landfill site is plotted in Figure 6.7. It decreases from  $2.909\text{E-}10 \text{ mg/m}^3$  to  $9.1\text{E-}11 \text{ mg/m}^3$  during 9 years. They are far less than  $1\text{mg/m}^3$  of the reference concentration (USEPA 1999a). Thus, it is in a very safe condition.

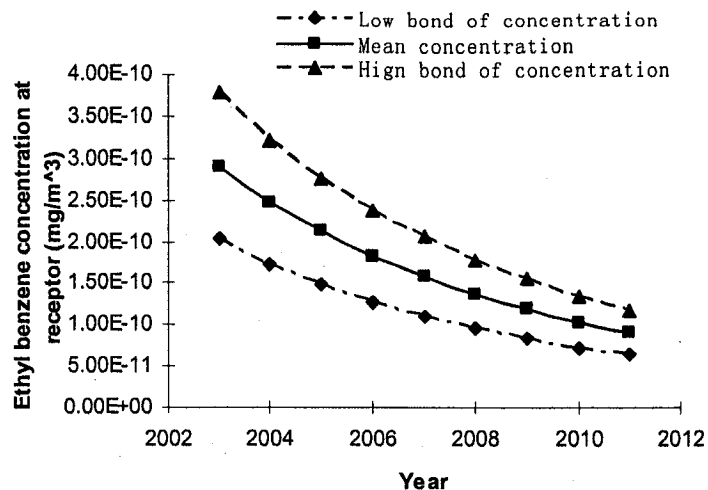


Figure 6.7 Yearly ethylbenzene profile at ground surface receptor for 2003-2011

Figure 6.8 shows the average concentration profile for toluene in the ambient air downwind from the landfill. It decreases sharply from  $3.773\text{E-}10 \text{ mg/m}^3$  to  $8.73\text{E-}11 \text{ mg/m}^3$ , which is far less than the  $5 \text{ mg/m}^3$  of the reference concentration (USEPA 2005).

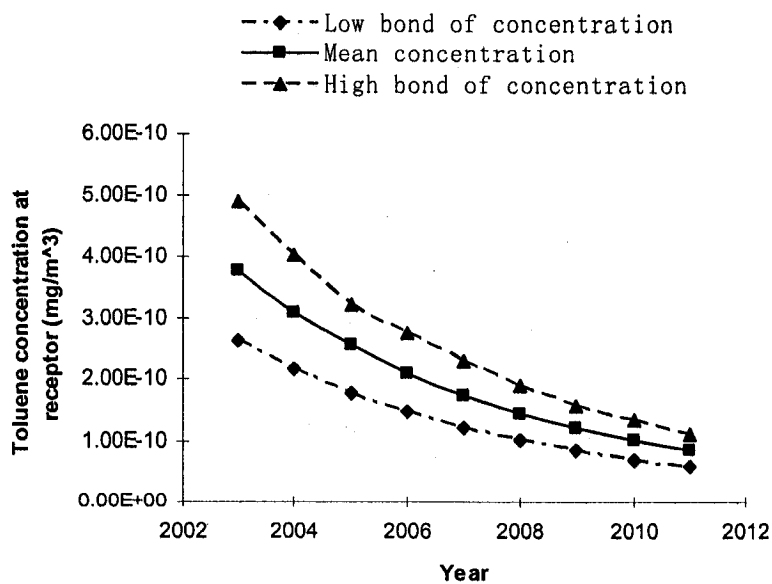


Figure 6.8 Yearly toluene profiles at ground surface receptor for 2003-2011

This emission flux is mainly attributed to the combined effect of Henry's law constant and the diffusion coefficient in the gas flux. The former characterizes the volatility of the contaminant, while the latter determines the emission amount through the landfill cover. Of the three contaminants, ethylbenzene has the largest Henry's law constant but the smallest diffusion coefficient, whereas benzene and toluene have almost the same Henry's law constants and diffusion coefficients. Thus, under the combined effect of these two parameters, the emission fluxes of toluene and ethylbenzene fall in a similar range.

#### **6.2.2.2 Predicted uncertainties**

The computed yearly variance of the benzene concentration at the receptor is given in the form of a fuzzy set output in Table 6.4, and that of concentration are plotted in Figure 6.6. The high bond is from  $3.889\text{E-}11 \text{ mg/m}^3$  to  $1.07\text{E-}11 \text{ mg/m}^3$ , and the low bond ranges from  $2.049\text{E-}11 \text{ mg/m}^3$  to  $5.76\text{E-}12 \text{ mg/m}^3$  for 2003-2011. Even the high bond of the simulated benzene concentration for every evaluated year at the exposure site is far less than the  $8.0\text{E-}2 \text{ mg/m}^3$  of the risk assessment reference concentration (USEPA 1999a, 2002). Thus, it can be concluded that the risk resulting from the inhalation of benzene for human beings at the exposure site can be neglected.

Table 6.4 Yearly benzene concentrations at ground surface receptor for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at receptor (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	209.4E-13	299.1E-13	388.9E-13
2004	175.5E-13	250.7E-13	325.9E-13
2005	148.0E-13	211.4E-13	274.8E-13
2006	125.4E-13	179.1E-13	232.8E-13
2007	106.6E-13	152.3E-13	198.0E-13
2008	91.0E-13	130.0E-13	169.1E-13
2009	77.9E-13	111.4E-13	144.8E-13
2010	66.9E-13	95.6E-13	124.3E-13
2011	57.6E-13	82.3E-13	107.0E-13

The fuzzy modelling results of ethylbenzene are expressed in Table 6.5 and the yearly concentration profile is shown in Figure 6.7. The fuzzy outputs in 2003 are 2.036E-10 mg/m<sup>3</sup>, 3.872E-10 mg/m<sup>3</sup>, and those in 2011 are 6.37E-11 mg/m<sup>3</sup>, and 1.183E-10 mg/m<sup>3</sup>. According to the 1mg/m<sup>3</sup> of the reference concentration (USEPA 1999a), the ethylbenzene has no adverse impact on human health at the exposure site.

Table 6.5 Yearly ethylbenzene concentrations at ground surface receptor for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at receptor (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	203.6E-12	290.9E-12	378.2E-12
2004	173.4E-12	247.6E-12	321.9E-12
2005	148.5E-12	212.1E-12	275.7E-12
2006	127.8E-12	182.5E-12	237.3E-12
2007	110.4E-12	157.8E-12	205.1E-12
2008	95.8E-12	136.9E-12	177.9E-12
2009	83.4E-12	119.1E-12	154.9E-12
2010	72.8E-12	104.0E-12	135.2E-12
2011	63.7E-12	91.0E-12	118.3E-12



The toluene concentrations for each evaluated year are displayed in Table 6.6. Figure 6.8 shows that the low bound of estimated concentration decreases considerably from  $2.641\text{E-}10 \text{ mg/m}^3$  to  $6.11\text{E-}11 \text{ mg/m}^3$  and the upper limit decreases from  $4.905\text{E-}10 \text{ mg/m}^3$  to  $1.135\text{E-}10 \text{ mg/m}^3$ . The toluene concentrations at the assessment receptor are much smaller than  $5 \text{ mg/m}^3$  of the reference concentration (USEPA 2005) so that the risk impact of toluene on human health is negligible.

Table 6.6 Yearly toluene concentrations at ground surface receptor for 2003-2011

Year	Low bound of concentration (mg/m <sup>3</sup> )	Modelled concentration at receptor (mg/m <sup>3</sup> )	High bound of concentrations (mg/m <sup>3</sup> )
2003	264.1E-12	377.3E-12	490.5E-12
2004	216.8E-12	309.7E-12	402.6E-12
2005	178.9E-12	255.5E-12	322.2E-12
2006	148.3E-12	211.8E-12	275.3E-12
2007	123.4E-12	176.3E-12	229.1E-12
2008	103.0E-12	147.2E-12	191.4E-12
2009	86.3E-12	123.3E-12	160.4E-12
2010	72.6E-12	103.7E-12	134.8E-12
2011	61.1E-12	87.3E-12	113.5E-12

### 6.3 Summary

A user-friendly GUI is designed for the developed EMMS. All the required input parameters are arranged in six input categories, making the interface clear and easy to understand.

EMMS and the fuzzy-set enhanced EMMS are applied to the Trail Road Landfill to simulate BET concentration profiles in the groundwater and the air from 2003 to 2011

and to assess the potential risk to humans of exposure to BET. In general, some conclusions can be obtained from the EMMS execution for future predictions:

1. Contaminant gas diffusive flux and dissolution in leachate are dominant mechanisms for the landfill of 6-year age. The landfill cover with high density prevents the emission of a contaminant gas from a landfill surface. Therefore, the emission flux from the landfill is a minor portion of contaminant release from a landfill.

2. The selection of input parameters has significant effects on the outputs of the modelling because of the uncertainties in the model formulation and in input parameters. The application of a fuzzy set to modelling inputs can handle these uncertainties with a fuzzy output and can give a more effective decision space for environmental management.

3. The quality and availability of field data influences the modelling outputs. This has also been reasonably handled by the fuzzy-set approach.

## **Chapter 7**

### **Conclusion and Future Study**

#### **7.1 Conclusion**

A new environmental multimedia modelling system (EMMS-PRO) has been developed in this present thesis study, which is intended to address the risks that arise from a contamination site with multimedia impacts. The modelling system is composed of four key modules: the polluting source (landfill) module, the unsaturated zone module, the groundwater module, and the air dispersion module. Critical improvements over previous EMMS are achieved by dynamically quantifying the intermedia mass flux and by thus enabling the developed EMMS to address complex multimedia environmental problems. In addition to this consideration, individual module components are thoroughly designed as solid model components capable of dealing with single medium pollution risks.

The developed EMMS-PRO has been systematically tested and validated by using a series of case studies and comparisons with literature data and field observations. Both the verification studies and the validation studies conducted in this present thesis

study indicate that the developed EMMS-PRO is a solid, versatile, and useful environmental multimedia risk assessment tool.

In order to make the developed system more useful and applicable, an effective uncertainty quantification technique based on fuzzy-set theory has been embedded in key modules and parameters based on an engineering consideration. It has been proved that this enhancement significantly helps the developed EMMS-PRO to deal with a huge amount of various uncertainties associated with environmental multimedia modelling practices.

Finally, a user-friendly GUI has been developed for EMMS-PRO to facilitate technology transfer and to provide significant help for the processing of model input and output.

## **7.2 Research Contributions**

In addition to the conclusions in section 7.1, the research contributions for this present thesis study are summarized below:

1. A new user-friendly fuzzy-set enhanced environmental multimedia modelling system (EMMS-PRO) has been developed.
2. The developed EMMS is grounded on the extension of previous efforts in single-medium or single-system modelling. Especially, the technical considerations of inter-media mass transfers or fluxes between air, the pollution source zone, soil, and groundwater are systematically considered in the first time. This makes of the developed

EMMS-PRO a unique risk assessment tool to address complex multimedia environmental pollution problems.

3. Two important enhancements have been made to enable the developed EMMS-PRO to be more useful and applicable. These enhancements are an innovative use of fuzzy-set theory to quantify various uncertainties and a user-friendly engineering interface.

4. The developed system has been systematically tested and validated, showing that EMMS-PRO can contribute extensively to environmental risk assessment and to effective pollution control decisions.

### **7.3 Recommendation for Future Studies**

Future studies that can be recommended are the following:

1. This study is the first engineering attempt in this field, part of dynamics including mass transfer and the flow involved are considered under equilibrium and steady-state conditions. Future study can include non-equilibrium and non-steady-state conditions.

2. Diffusion coefficients in the gas and liquid phases in a pollution source module and diffusion coefficients in soil and groundwater are estimated by using empirical equations that are derived from particular situations. Thus, this estimation may contribute to certain imprecision for different field conditions. It is recommended that more methods be examined.

3. Seepage velocities in an unsaturated zone and a saturated zone are estimated using Darcy's law and are regarded as constant during the evaluation period. In reality, however, these velocities will deviate from this assumption and vary from time to time. In future studies, the variable seepage velocity in the aquifer zone can be considered and simulated by integrating sub-models characterizing the seepage velocity as the function of the meteorological conditions.

4. The effects of infiltration have been studied in the present thesis. However, the impact of precipitation needs to be further considered in the future. That will involve more hydrological input or a hydrological module.

5. In present uncertainty analysis, the quantification of uncertainties is obtained from iterative simulations. These results can be combined with sensitive analysis to understand more about important model uncertainties.

## References

- Allen, D.T., Cohen, Y., and Kaplan, I.R., 1989. *Intermedia Pollutant Transport: Modelling and Field Measurements*. New York: Plenum Publisher.
- Ayra, A., 1986. *Dispersion and reservoir heterogeneity*, Ph.D. Dissertation, University of Texas, Austin, TX.
- Berg, M. and Scheringer, M., 1994. Problems in environmental risk assessment and the need for proxy measures, *Fresenius Environmental Bulletin*, 3: 486-492.
- Berry-spark, K., Barker, J.F., Major, D., and Mayfield, C.I., 1986. Bioremediation of gasoline-contaminated ground-waters: A controlled field experiment. In proceedings, *petroleum Hydrocarbons and Organic Chemicals in Groundwater: Prevention, Detection and Bio restoration*, NWWA/API: Houston, TX, 613-623.
- Bouwer, E.J. and McCarty, P.L., 1983. Kinetics of secondary substrate utilization. *National Meeting - American Chemical Society*, Division of Environmental Chemistry, 23(2): 448-452.
- Carnahan, C.L. and Remer, J.S., 1984. Nonequilibrium and Equilibrium sorption with a linear sorption isotherm during mass transport through an infinite porous medium: some analytical solution. *Journal of Hydrology*, 73: 227-258.
- Cohen, Y., 1986. *Pollutants in a multimedia environment*. Plenum press, New York.

- Cohen, Y. and Ryan, P.A., 1989. Chemical transport in the top soil zone-the role of moisture and temperature gradients. *Journal of Hazardous Materials*, 22: 283-304.
- Cohen, Y. and Ryan, P.A., 1985, Multimedia Modelling of Environmental Transport: Trichloroethylene Test Case, *Environmental Science and Technology*, 9: 412-417.
- Cohen, Y., Tsai, W., Chetty, S.L., and Mayer, G.J., 1990. Dynamic Partitioning of Organic Chemicals in Regional Environments: A Multimedia Screening-Level Approach, *Environmental Science and Technology*, 24: 1549-1558.
- Cohen, Y. and Cooter, E., 2002. Multimedia environmental distribution of Toxics (Mend-Tox) I: hybrid compartment-spatial modelling framework. *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, 6(2): 70-86.
- Cooper, C.A., Reinhart, D.R., Rash, F., and Seligman, D.K., 1992. *Landfill gas emission*. Technical Report. Civil and Environmental Engineering Department University of Central Florida, Florida, Centre for Solid and Hazardous Waste Management.
- Cowan, C.E., Mackay, D., Feijtel, T.C.J., Van De Meent, D., and Di Guardo, A., 1995. *The Multi-Media Fate Model: A Vital Tool for Predicting the Fate of Chemicals*. Pensacola, FL: Society of Environmental Toxicology and Chemistry.



- den Hollander, H.A. and van de Meent, D., 2004. *SimpleBox 3.0: A multimedia mass balance model for evaluating the environmental fate of chemicals*. National Institute of Public Health and the Environment, Bilthoven, the Netherlands.
- Devillers, J., Bintein, S., and Karcher, W., 1995. CHEMFRANCE: A regional level III fugacity model applied to France. *Chemosphere*, 30(3): 457-476.
- Domenico, P.A., 1987. An analytical model for multidimensional transport of a decaying contaminant species. *Journal of Hydrology*, 91: 49-58.
- Droppo, J.G., Streng, D.L., Buck, J.W., Hoopes, B.L., Brockhaus, R.D., Walter, M.B., and Whelan, G., 1989. *Multimedia Environmental Pollutant Assessment System (MEPAS) Application Guidance Volume 1-User's Guide*. Pacific Northwest Laboratory, Richland, WA.
- Droppo, J.G., Buck, J.W., Streng, D.L., and Hoopes, B.L., 1993. Risk computation for environmental restoration activities. *Journal of Hazardous Materials*, 35: 341-352.
- Dubois, D. and Prade, H., 1980. *Fuzzy Sets and Systems: Theory and Application*. Mathematics in science and Engineering, Volume 144.
- Eisenbud, M., 1987. *Environmental Radioactivity from Natural, Industrial, and Military Sources*. 3rd ed. New York: Academic.
- Evans, G.W., Karwowski, W., and Wilhelm, M.R., 1989. An introduction to fuzzy set methodologies for industrial and system engineering. *Applications of Fuzzy Set Methodologies in Industrial Engineering*, edited by Evans, G.W., Karwowski, W., and Wilhelm, M.R., ELSEVIER.

- Fetter, C. W., 1993. *Contaminant Hydrogeology*. Macmillan Pub. Co.
- Garrels, R.M., Mackenzie, F.T., and Hunt, C., 1975. *Chemical Cycles and the Global Environment: Assessing Human Influences*. Los Altos, CA: Kaufmann.
- Gelhar, L. W., Welty, C., and Rehfeldt, K. R., 1992. A critical review of data on field-scale dispersion in aquifers. *Water Resources Research*, 28(7): 1955-1974.
- Hsieh, C.R. and Ouimet, J.R., 1994. Comparative study of multimedia modelling for dynamic partitioning of fossil fuels-related pollutants. *Journal of Hazardous Materials*, 37: 489-505.
- Hodson, J. and Williams N.A., 1988. The estimation of the adsorption coefficient (K<sub>oc</sub>) for soils by high performance liquid chromatography. *Chemosphere*, 17: 67-77.
- Hogdson, A.T., Garbesi, K., Sextro, R.G., and Daisry, J.M., 1992. Soil-gas contamination and entry of volatile organic compounds into house near a landfill. *Journal of Air waster Management Association*, 42: 277-283.
- Howard, P.H., 1990. *Handbook of environmental fate and exposure data for organic chemicals*. Vol. II. Solvents. Chelsea, MI: Lewis Publishers, Inc.
- Howard, P.H., Beothling, R.S., Jarvis, W.F., Meylan, W.M., and Michalenko, E.M., 1991. *Handbook of Environmental Degradation Rates*. Chelsea, MI: Lewis Publishers, Inc.
- HSDB, 2005. *Benzene, Hazardous Substances Data Bank*. National Library of Medicine. <http://toxnet.nlm.nih.gov>.

- Hutchins, S.R., Sewell, G.W., Kovacs, D.A., and Smith, G.A., 1991a. Biodegradation of aromatic hydrocarbons by aquifer microorganisms under denitrifying conditions. *Environmental Science and Technology*, 25: 68-76.
- Hutchins, S.R., Downs, W.C., Wilson, J.T., Smith, G.B., Kovacs, D.A., Fine, D.D., Douglass, R.H., and Hendrix, D.J., 1991b. Effect of nitrate addition on biotransformation of fuel-contaminated aquifer: Field demonstration. *Ground Water*, 29(4): 571-580.
- IAEA, 1989. *Evaluating the reliability of predictions made using environmental transfer models*. International Atomic Energy Agency. Safety Series No.100, Vienna, Austria.
- Inuiguchi, M., Ramik, J., 2000. Possibilistic linear programming: a brief review of fuzzy mathematical programming and a comparison with stochastic programming in portfolio selection problem. *Fuzzy Sets and System*, 111:3-28.
- Jahn, M.K., Haderlein, S.B., and Meckenstock, R.U., 2005. Anaerobic degradation of benzene, toluene, ethylbenzene, and o-xylene in sediment-free iron-reducing enrichment cultures. *Applied and Environmental Microbiology*, 71(6): 3353-3358.
- Jury, W.A., Russo, D, Strelie, G. and Abd, H.E., 1990. Evaluation of volatilisation by organic chemicals residing below the soil surface. *Water Resource Research*, 26(1): 13-20.

- Jury, W.A., Spencer, W.F., and Farmer, W.J., 1983. Behaviour assessment model for trace organics in soil: I. model description. *Journal of Environmental Quality*, 12(4): 558-564.
- Kaufmann, A., Gupta, M. M., 1985. *Introduction to Fuzzy Arithmetic Theory and Application*. Van Nostrand Reinhold Company, New York.
- Khim, J., 1994. *Uncertainty analysis of VOC emissions models for closed landfills and land treatment sites*. Ph. D. dissertation. The University of Texas at Austin.
- Kuhn, E.P., Zeyer, J., Eicher, P., and Schwarzenbach, R.P., 1988. Anaerobic degradation of alkylated benzenes in denitrifying laboratory aquifer columns. *Applied Environmental Microbiology*, 54: 490-496.
- Labieniec, P.A., 1994. *The risk implications of approaches for setting soil remediation goals at hazardous waste contaminated site*. PhD thesis, Carnegie Mellon University, Pittsburgh, Pa.
- Labieniec, P.A., Dzombak, D.A., and Siegrist, R.L., 1996a. Soil risk: risk assessment model for organic contaminants in soil. *Journal of Environmental Engineering*, 122: 388-398.
- Labieniec, P.A., Dzombak, D.A., and Siegrist, R.L., 1996b. Risk variability due to uniform soil remediation goals. *Journal of Environmental Engineering*, 122(7): 612-621.
- Laniak, G.F., Droppo, J.G., Faillace, E.R., and Gnanapragasam, E.K., 1997. An overview of a multimedia benchmarking analysis for three risk assessment model: RESRAD, MMSOILS, and MEPAS. *Risk analysis*, 17(2): 203-214.

- Lin, J. S. and Hildemann, L.M., 1995. A non-steady-state analytical model to predict gaseous emissions of volatile organic compounds from landfills. *Journal of Hazardous Materials*, 40: 271-295.
- Liu, L., Cheng, S.Y., and Guo, H.C., 2004. A simulation-assessment modelling approach for analysing environmental risks of groundwater contamination at waste landfill sites. *Human and Ecological risk assessment*, 10: 373-388.
- Lyman, W.J., Reehl, W.F., Rosenblatt, D.H., 1990. *Handbook of chemical property estimation methods*. Washington, DC: American Chemical Society.
- Mackay, D. and Shiu, W.Y., 1981. A critical review of Henry's law constants for chemicals of environmental interest. *Journal of Physical and Chemical Reference Data*, 19: 1175-1199.
- Mackay, D., Shiu, W.Y., and Sutherland, R.P., 1979. Determination of air-water Henry's law constants for hydrophobic pollutants. *Environmental Science Technology*, 13: 333-337.
- Mackay, D. and Leinonen, P.J., 1975. Rate of evaporation of low-solubility contaminants from water bodies to atmosphere. *Environmental Science Technology*, 9: 1178-1180.
- Mackay, D., 1979. Finding fugacity feasible. *Environmental Science Technology*, 13: 1218-1223.
- Mackay, D. and Paterson, S., 1981. Calculating fugacity. *Environmental Science Technology*, 15: 1006-1014.

- Mackay, D. and Paterson, S., 1982. Fugacity revisited—the fugacity approach to environmental transport. *Environmental Science Technology*, 16: A654–660.
- Mackay, D., 1991. *Multimedia Environmental Models: The Fugacity Approach*. Lewis Publishers, Chelsea, Mich.
- Mackay, D., Paterson, S., and Tam, D.D., 1991. *Assessments of Chemical Fate in Canada: Continued Development of a Fugacity Model*. A report prepared for Health and Welfare Canada.
- MacLeod, M., Woodfine, D.G., Mackay, D., McKone, T., Bennet, D., and Maddalena, R., 2001. BETR North America: a regionally segmented multimedia contaminant fate model for North America. *Environmental Science and Pollutant Research*, 8 (3): 156-163.
- Maddalena, R.L. and McKone, T.E., 1995. Comparison of multimedia transport and transformation models: regional fugacity models vs. CalTOX. *Chemosphere*, 30(5): 869-889.
- Majora, D.W., Mayfield, C.I., and Barkerb, J.F., 1988. Biotransformation of benzene by denitrification in aquifer sand. *Groundwater*, 26: 8-14.
- McDonald, J.P., and Gelston, G.M., 1998. Description of the multimedia environmental pollutant assessment system (MEPAS, Version 3.2), with application to a hypothetical soil contamination scenario. *Journal of Soil Contaminant*, 7(3): 283-300.

- McKone, T.E. and Enoch, K.G., 2002. CalTOX, *A multimedia total exposure model spreadsheet user's guide Version 4.0*. Ernest Orlando Lawrence Berkeley National Laboratory.
- McKone, T.E. and MacLeod, M., 2003. Tracking multiple pathways of human exposure to persistent multimedia pollutants: regional, continental, and global-scale models. *Annual Review of Environment and Resources*, 28: 463-492.
- Mills, W.B., Cheng, J.J., Droppo, J.G., and Faillace, E.R., 1997. Multimedia benchmarking analysis for three risk assessment models: RESRARD, MMSOIL, and MEPAS. *Risk analysis*, 17(2): 187-201.
- Millington, R.J., and Quirk, J.P., 1961. Permeability of porous solids. *Transactions of the Faraday Society*, 57(7): 1200-1207.
- Nastev, M., 1998. *Modelling landfill gas generation and migration in sanitary landfills and geological formations*. Ph. D. thesis. Laval University.
- Ng, Y.C., 1982. A review of transfer factors for assessing the dose from radionuclides in agricultural products. *Nuclear Safety*. 23: 57-71.
- Nriagu, J.O., 1978. *The Biogeochemistry of Lead in the Environment*. New York: Elsevier/North-Holland Biomed.
- Nriagu, J.O., 1979. *The Biogeochemistry of Mercury in the Environment*. New York: Elsevier/North-Holland Biomed. Chelsea, MI: Lewis.
- Reinhart, D.R., Cooper, C.D., and Ruiz, N.E., 1994. *Estimation of Landfill Gas Emissions at the Orange County Landfill*, Civil and Environmental Engineering Department, University of Central Florida, Orlando.

- Rickabaugh, J.F., 1990. *Evaluation of Trace VOC Emissions from Sanitary Landfills*. Ph.D. dissertation. Department of Civil and Environmental Engineering of the College of Engineering, University of Cincinnati.
- Scheringer, M., 1996. Persistence and spatial range as endpoints of an exposure-based assessment of organic chemicals. *Environmental Science and Technology*, 30: 1652–1659.
- Scheringer, M., 1997. Characterization of the environmental distribution behavior of organic chemicals by means of persistence and spatial range. *Environmental Science and Technology*, 31: 2891–2897.
- Scheringer, M., Stroebe, M., and Held, H., 2002. *Chemrange 2.1- a multimedia transport model for calculating persistence and spatial range of organic chemicals*. Swiss Federal Institute of Technology Zurich.
- Solhotra, A.M., Mineart, P., Sharp-Hansen, S., and Allison, T.L., 1990. *Multimedia Exposure Assessment Model (MULTIMED 1.5) for evaluating the land disposal of wastes—model theory*. Prepared for office of research and development, U.S. Environmental Protection Agency, Athens, Georgia.
- Solhotra, A.M., Mineart, P., Sharp-Hansen, S., Allison, T., John, R., and Mills, W.B., 1995. *Multimedia exposure assessment model (MULTIMED 2.0) for evaluating the land disposal of waste-model theory*. Environmental Research Lab, U.S. EPA, Athens, Ga.
- Streile, G., 1988. *Unpublished notes providing derivations of equations in Jury et al. (1983, 1990)*. Batelle Pacific northwest Laboratory, Richland, Wash.



Strand, S. *Anaerobic Degradation of Aromatic Compounds: ring Cleavage without Oxygen*. Lectures notes, College of Forest Resources University of Washington.

The Council on Environmental Quality (TCEO), 1985. *Report on Long-Term Environmental Research and Development*, Executive Office Press. US, CEQ, Washington, DC.

Tchobanoglous, G., Theisen, H., and Vigil, S., 1993. *Integrated Solid Waste Management*. McGraw-Hill, New York.

Thibodeaux, L.J., 1981. Estimating air emissions of chemicals from hazardous waste landfills. *Journal of Hazardous Materials*, 4(3): 235-244

Thibodeaux, L.J., Springer, C., and Riley, L.M., 1981. Models of mechanisms for hazardous chemical emissions from landfills. Presented at the Symposium on Toxic Substances Management Programs, *181th American Chemical Society National Meeting*, Atlanta, GA, 1981.

Thibodeaux, L.J., Springer, C. and Riley, L.M., 1982. Models of mechanisms for vapour phase emission of hazardous chemicals from landfills. *Journal of hazardous Materials*, 7(1): 63-74.

TRNL, 1995. *Trail Road and Nepean Landfill Sites Final Report for the 1995 Monitoring and Operation Program*. Prepared by M.M.Dillon Limited and Gartner Lee Limited. Environment and Transportation Department Solid Waste Division.

- TRNL, 2002. *Trail Road and Nepean Landfill Sites Final Report for the 2002 Monitoring and Operation Program*. Prepared by M.M.Dillon Limited and Gartner Lee Limited. Environment and Transportation Department Solid Waste Division.
- Turner, D.B., 1970. *Workbook of atmosphere dispersion estimates*. U.S. Department of Health, Education, and Welfare, Washington, D.C.
- USEPA, 1986. *Development of advisory levels for PCB cleanup*. Office of Health and Environmental Assessment, Washington, D.C.
- USEPA, 1988. *Methodology for estimating multimedia exposures to soil contamination (MMSOIL)*. Office of Health and Environmental Assessment, Exposure Assessment Group, Washington, D.C.
- USEPA, 1996. *Three multimedia models used at hazardous and radioactive waste sites*. U.S. Environmental Protection Agency, Washington, D.C.
- USEPA, 1999a. *Integrated Risk Information System (IRIS) on Ethylbenzene*. National Center for Environmental Assessment, Office of Research and Development, Washington, DC.
- USEPA, 1999b. *FRAMES-HWIR: Technology software system for 1999, Vol, I, system overview*. U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C. (Draft July).
- USEPA, 2002. *Toxicological review of benzene (noncancer effects)*. In Support of Summary Information on the Integrated Risk Information System (IRIS). U.S. Environmental Protection Agency. Washington D.C.

- USEPA, 2003. *SAB Review Materials for 3MRA Modeling System Volume I: Modeling System and Science*. U.S. Environmental Protection Agency, Washington, D.C.
- USEPA, 2005. *Toxicological review of Toluene. In Support of Summary Information on the Integrated Risk Information System (IRIS)*. U.S. Environmental Protection Agency. Washington D.C.
- van der Heijde, P.K.M.. 1987. Quality assurance in computer simulations of ground water contamination. *Environmental Software*, 2(1): 19-25.
- Webster, E., Mackay, D., Di Guardo, A., Kane, D., and Woodfine, D., 2004. Regional differences in chemical fate model outcome. *Chemosphere*, 55: 1361-1376.
- Wexler, E.J., 1992. *Analytical solutions for one-, two-, and three-dimensional solute transport in ground-water systems with uniform flow*. U.S. Geological Survey, Denver.
- Whicker, F.W. and Kirchner, T.B., 1987. Pathway: a dynamic food-chain model to predict radionuclide ingestion after fallout deposition. *Health Physics*, 52: 717-737.
- Wilson, J.L., Conrad, S.H., Mason, W.R., Peplinski, W., and Hagan, E., 1990, *Laboratory Investigation of Residual Liquid Organics*, United States Environmental Protection Agency, EPA.600/6-90/004.
- Xu, M. and Eckstein, Y., 1995. Use of weighted least-squares method in evaluation of the relationship between dispersivity and field scale, *Groundwater*, 33(6): 905-908.
- Yalkowsky, S.H. and Valvani, S.C., 1976. Partition coefficients and surface areas of some alkylbenzenes. *Journal of Medicinal Chemistry*, 19: 727-728.

Zadeh, L.A., 1973. Outline of a new approach to the analysis of complex systems and decision processes. *IEEE transactions on System, Man and Cybernetics*, 3(1): 28-44.

Zeyer, J., Kuhn, E.P., and Schwarzenbach, R.P., 1986. Rapid Microbial Mineralization of Toluene and 1,3-Dimethylbenzene in the Absence of Molecular Oxygen. *Applied Environmental Microbiology*, 52: 944-947.

Zimmermann, H.J., 2001. *Fuzzy Set Theory and Its Applications*. Kluwer Academic Publishers, Boston/Dordrecht/London.

Spectrum Laboratories online datasheet:

<http://www.speclab.com/compound/c108883.htm>

The Weather Network:

<http://www.theweathernetwork.com/weather/cities/can/Pages/CAON0512.htm>

# Appendix

## Derivation of Equation [3.38]

$$\frac{dM(t)}{dt} = m_0 \exp[-(\gamma + \mu)t] - \mu M(t) \quad [3.39]$$

The Laplace transform of Equation [3.39] is:

$$sM(s) - M(0) = \frac{m_0}{s + \gamma + \mu} - \mu M(s) \quad [A.1]$$

where  $s$  is Laplace transform variable.

With the initial condition  $M(0)=0$

Equation [A.1] can be in the form of:

$$M(s) = \frac{m_0}{(s + \mu)(s + \mu + \gamma)} \quad [A.2]$$

In order to solve Equation [A.2], we change it to be:

$$M(s) = \frac{m_0}{\gamma} \left( \frac{1}{s + \mu} - \frac{1}{s + \mu + \gamma} \right) \quad [A.3]$$

Inverting Equation [A.3], the solution to Equation [3.39] can be obtained:

$$M(t) = \frac{m_0}{\gamma} [\exp(-\mu t) - \exp(-\mu - \gamma)t]$$

which is Equation [3.38].