3-2005

Validation of an Innovative Groundwater Contaminant Flux Measurement Method

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VALIDATION OF AN INNOVATIVE GROUNDWATER CONTAMINANT FLUX MEASUREMENT METHOD

THESIS

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AFIT/GES/ENV/05M-02

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THESIS

Presented to the Faculty

Department of Systems and Engineering Management

Graduate School of Engineering and Management

Air Force Institute of Technology

Air University

Air Education and Training Command

In Partial Fulfillment of the Requirements for the

Degree of Master of Science in Environmental Engineering and Science

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March 2005

AFIT/GES/ENV/05M-02

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Abstract

The ability to measure groundwater contaminant flux is increasingly being recognized as crucial in order to prioritize contaminated site cleanups, estimate the efficiency of remediation technologies, measure rates of natural attenuation, and apply proper source terms to model groundwater contaminant transport. An innovative mass flux measurement method using horizontal flow treatment wells (HFTWs) was developed recently to compensate for the disadvantages of other flux measurement methods that are being used.

Flux measurement methods can be categorized as either point methods or integral methods. As the name suggests, point methods measure flux at a specific point or points in the subsurface. To increase confidence in the accuracy of the measurement, it is necessary to increase the number of points (and therefore, the cost) of the sampling network. Integral methods avoid this disadvantage by using pumping wells to interrogate large volumes of the subsurface. Unfortunately, integral methods are expensive because they require that large volumes of contaminated water be extracted and managed. HFTWs combine the advantages of each of the two approaches described above; that is, it¡¯s an integral technique that samples a large volume of the subsurface while not requiring extraction of contaminated water from the subsurface.

In this study, the accuracy of the HFTW flux measurement method was quantified by applying the method in an artificial aquifer, where the flux being measured was known. Two HFTW approaches, the multi-dipole approach and the tracer test approach, were

compared to each other, as well as being compared to the transect method of measuring flux, which is the conventionally used point method.

Results found that the transect and HFTW tracer test approaches provided reasonably accurate measures of flux (within ±50% and ±44% respectively) in the artificial aquifer, while the multi-dipole approach was too sensitive to small hydraulic head measurement errors to be useful. A comparison of the costs of applying the different methods at a generic site showed that the HFTW method had significant cost advantages. This study also compared other advantages and disadvantages of the various flux measurement methods, concluding that depending on conditions at a site, one or the other method may be most advantageous for application.

Acknowledgments

First and foremost, I would like to express my deepest gratitude to my thesis advisor, Dr. Mark N. Goltz, for taking the challenge of guiding me through this thesis effort. Without his guidance and endless support, I would not have dreamed of coming to such a state of accomplishment. The experience, insight and knowledge which he has shared with me are invaluable and well appreciated. I would also like to thank Dr. Alfred E. Thal Jr. and Dr. Junqi Huang for their expertise and guidance which were great assets in writing my thesis.

My sincere thanks to Murray Close, Mark Flintoft, and their colleagues at the New Zealand Institute of Environmental Science and Research and AquaLinc Research Ltd. for operating the artificial aquifer and providing the data used in this study.

This study was partially supported by the Environmental Security Technology Certification Program (ESTCP) through Project CU-0318, Diagnostic Tools for Performance Evaluation of Innovative In-Situ Remediation Technologies at Chlorinated Solvent-Contaminated Sites. Also, I acknowledge the Air Force Center for Environmental Excellence (AFCEE) who sponsored this research.

In addition, I am indebted to the Korean Army for providing me with this wonderful opportunity of Master s degree program that has broadened my knowledge as well as stimulated my understanding of military technology. Hopefully, all this will help me better serve my country in the future.

Most importantly, I am forever thankful to my wife and son for being there with love, support and understanding. I also feel grateful for my parents and in-laws in Korea for their support and encouragement. Thank you everyone for your patience, endurance, and strong belief in me. Seh Jong Kim

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VALIDATION OF AN INNOVATIVE GROUNDWATER CONTAMINANT FLUX MEASUREMENT METHOD

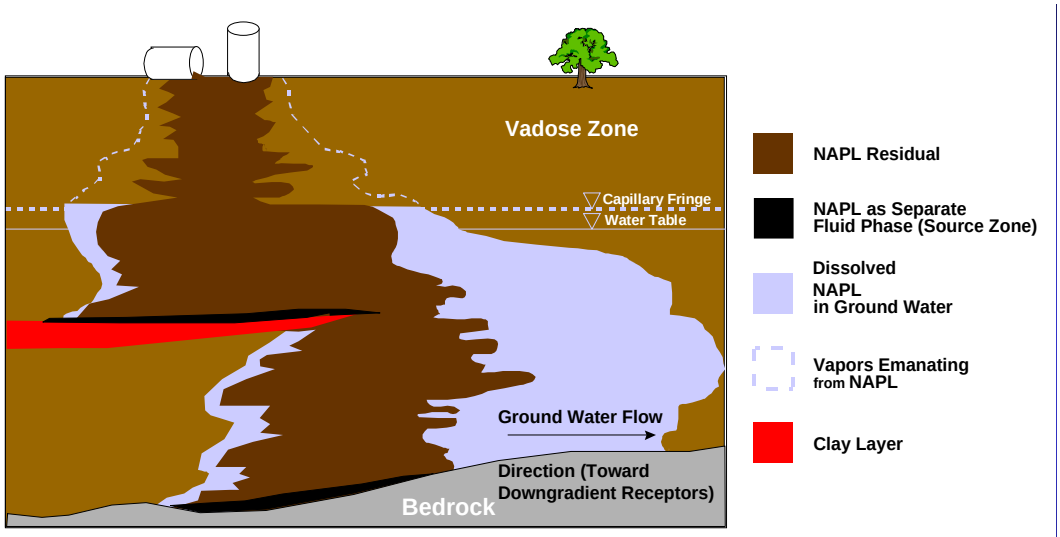
I. Introduction

1.1 Motivation

Groundwater constitutes about two thirds of the freshwater resources of the world and, if the polar ice caps and glaciers are not considered, groundwater accounts for nearly all usable freshwater (UNESO/WHO/UNEP, 1992). Even if consideration is limited to only the most active and accessible groundwater aquifers, then groundwater still makes up 95% of total freshwater, with lakes, swamps, reservoirs and rivers accounting for 3.5% and soil moisture accounting for only 1.5% (Freeze and Cherry, 1979). Groundwater has been extracted for domestic use (drinking, cleaning) as well as for agriculture (water for livestock and irrigation) since the earliest times. In the USA, where groundwater is important in all regions, about 40% of public water supplies overall rely on a groundwater source. In rural areas of the USA, 96% of domestic water is supplied from groundwater (UNESO/WHO/UNEP, 1992). Also, many of the major cities of Europe are dependent on groundwater.

At the same time that reliance on groundwater is growing throughout the world, groundwater resources are facing an unprecedented risk of contamination due to subsurface releases of chemicals (Einarson and Mackay, 2001). Contaminated

groundwater sites can be considered to consist of two parts, the source and the plume. Subsurface source areas typically are created when contaminants are either accidentally or intentionally released on or below the ground from drums, tank, landfills, etc. Many times these releases consist of contaminants such as oils and solvents that exist as separate phase liquids, commonly referred to as nonaqueous-phase liquids (NAPL), in the subsurface (Figure 1).



After NRC, 1994

Figure 1. **Groundwater contamination source zone and plume**

These separate phase contaminants migrate through the subsurface, moving by gravity through the vadose, or unsaturated zone until they reach the water table (Wiedemeier *et al.*, 1999). As the NAPL passes through the vadose zone, it leaves behind residual levels of pure phase contaminant, held between the grains of the porous media by capillary forces (Wiedemeier *et al.*, 1999). NAPLs that are less dense than

water, such as petroleum hydrocarbons, are called light-NAPLs (LNAPLs). LNAPLs will form a layer or pool that floats above the water table, slowly dissolving into groundwater passing below it. NAPLs such as chlorinated solvents are denser than water. These NAPLs, referred to as dense-NAPLs (DNAPLs), will sink below the water table, leaving behind residual droplets (see Figure 1). Eventually, the DNAPL will reach a low permeability layer, where it will spread out, creating a separate phase DNAPL pool (Wiedemeier *et al.*, 1999).

When released as a NAPL, large quantities of contaminants can be trapped in soils as residual droplets and pools. Due to the relatively low water solubility of many NAPL contaminants, the NAPL may persist for decades, only slowly dissolving into passing groundwater, to form contaminant plumes that can extend for miles (Einarson and Mackay, 2001). These plumes can ultimately be transported by flowing groundwater to receptors such as downgradient supply wells or surface water (Einarson and Mackay, 2001). In the United States alone, releases of gasoline fuels containing MTBE (methyl *tert*-butyl ether) may have occurred at more than 250,000 sites, with the potential to contaminate over 9000 large municipal water supply wells (Einarson and Mackay, 2001).

In 1980, the US government enacted the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) to address the risks posed by past releases of contaminants into soil and groundwater. CERCLA established a multi-decade/multi-billion dollar program to identify, characterize, and remediate contaminated sites.

Due to limited resources, an important component of the CERCLA process is prioritization of sites to be remediated based upon risk to human health and the environment (Einarson and Mackay, 2001). One parameter that is important in

quantifying risk is contaminant mass flux (SERDP/ESTCP, 2001; Einarson and Mackay, 2001; API report, 2003). Mass flux is a measure of the rate contaminant mass is transported, in units of mass per time per area of aquifer orthogonal to the direction of groundwater flow. Einarson and Mackay (2001) argued that contaminant mass flux is more relevant as an indicator of risk at a downgradient water supply well than contaminant concentration in the plume, even though most of our efforts to date have been focused on quantifying contaminant concentrations in the plume. Einarson and Mackay (2001) go on to suggest that contaminant mass flux measurements would be more useful than concentration measurements in helping regulators and remediation decision makers prioritize cleanup among numerous contaminant release sites.

In addition to helping assess risk in order to prioritize contaminated site cleanups, mass flux measurements can also be used to (1) quantify how readily a dissolved contaminant is degrading by natural processes (Borden *et al.*, 1997; Bockelmann *et al.*, 2003; Peter *et al.*, 2004), (2) evaluate the efficacy of cleanup technologies (SERDP/ESTCP, 2001; Soga *et al.*, 2002), and (3) determine the source term for use in contaminant transport modeling (Wiedemeier *et al.*, 1999). Contaminant flux measurement has been the subject of considerable research in the past five years, as scientists, regulators, and hazardous waste site managers have begun to realize the importance of measuring contaminant flux, as opposed to ¡°traditional¡± measurements o contaminant concentration (SERDP/ESTCP, 2002).

The conventional method of determining contaminant mass flux is to install a transect of multilevel sampling wells perpendicular to the direction of groundwater flow (the so-called transect method) (API, 2003) (Figure 2).

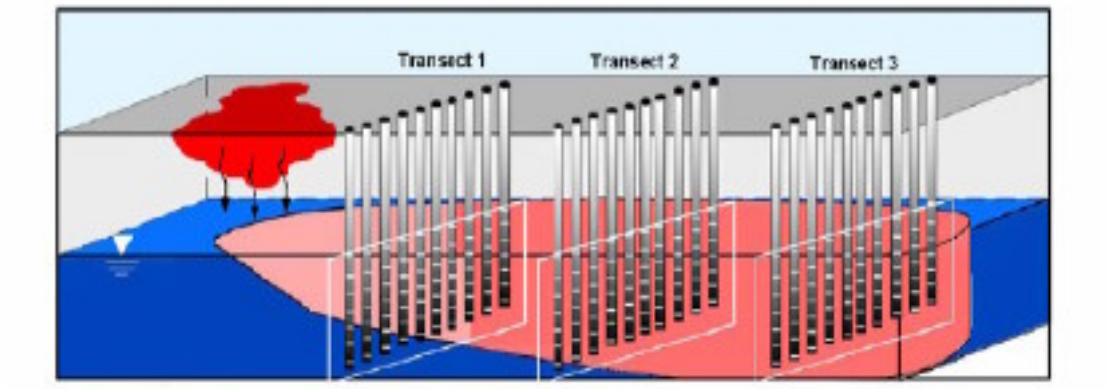


Figure 2. **Example of transect method of flux measurement using three control planes (API Groundwater Remediation Strategies Tool, 2003)**

This method may be categorized as a ¡°point method¡± of determining flux, in tha flux is measured at a number of sampling points. The disadvantage of point methods is due to the fact that sampling is at discrete points across the direction of flow. Thus, a large representative volume of the subsurface is not necessarily interrogated. Increasing the detail or range of sampling requires increasing the number (and therefore cost) of sampling wells.

Recently, the need for improved flux measurement techniques has led to the development of several innovative approaches. One new method that is currently being tested is a so-called ¡°integral approach¡± in that it involves pumping in order to integrat the flux measurement over the volume of contaminated groundwater that is pumped. This integral groundwater investigation method (IGIM) measures flux by operating one or more extraction wells installed along a plane perpendicular to the flow of groundwater (Bockelmann *et al.*, 2003) (Figure 3).

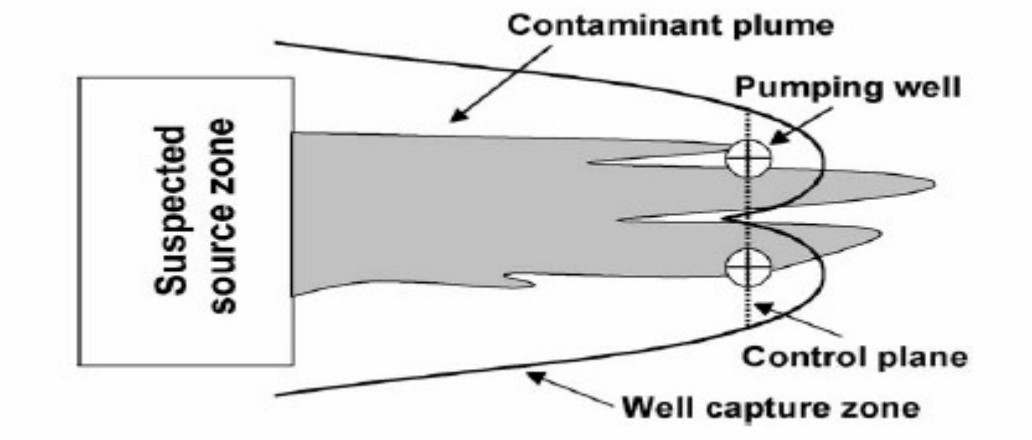


Figure 3. **Example of IGIM for flux measurement using two wells in one control plane (Bockelmann *et al.*, 2003)**

While the IGIM has the advantage of interrogating and averaging mass flux over a relatively large subsurface volume, the method incurs the expense of extracting and managing a large volume of contaminated water, as well as the increased risks to workers and others associated with implementing an aboveground technology to treat the contaminated water. An alternative innovative approach that is in development, which avoids these costs, involves use of a ¡®passive flux meter (PFM)¡¯ in a well borehol (Hatfield *et al.*, 2001). As a point method, however, the PFM technique has the same limitations as the conventional transect method.

A fourth flux measurement technique has been proposed that involves use of a pair of dual-screened pumping wells (also known as horizontal flow treatment wells, or HFTWs) to measure contaminant mass flux (Huang *et al.*, 2004). HFTWs consist of two wells, with each well having an injection and extraction screen (Figure 4).

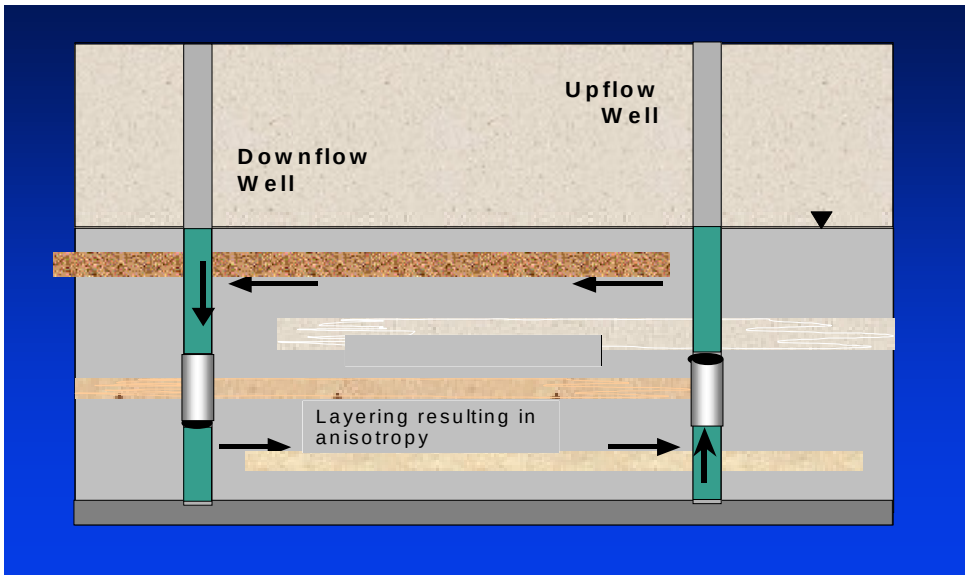


Figure 4. **Horizontal Flow Treatment Wells**

Water flows upwards in one well and downwards in the other. Note that water is never brought to the surface; it is just pumped from the extraction screen to the injection screen of a single well. Water injected into the aquifer through the injection screen then recirculates, flowing either to the extraction screen of the same well, the extraction screen of the second well, or flowing downgradient (Christ *et al.*, 1999). The proposed flux measurement technique using HFTWs combines the advantages of integral and point methods, while avoiding the disadvantages. That is, a large subsurface volume can be interrogated using the HFTW method without the need to extract large volumes of contaminated water (Huang *et al.*, 2004). While HFTWs have been applied in the field for contaminant plume cleanup (McCarty *et al.*, 1998), and HFTW flow models are available (Gandhi *et al.*, 2002), HFTWs have not been used in the past for flux measurement, although the theory for their use has been proposed by Goltz *et al.* (2004) and Huang *et al.* (2004).

Based on the need for improved methods of flux measurement, and the potential

of the HFTW technique to avoid the limitations of methods currently in use or under development, we propose to further study the HFTW technique. A crucial step in the development and commercialization of any new measurement technique is validation. Validation is defined as confirming an expected result as a true fact through reliable demonstration. In this case, we propose to validate the HFTW flux measurement technology by comparing the flux measured by the technique with a known flux. Validation is crucial if project managers, decision makers, and regulators are going to have confidence in the accuracy and reliability of the flux measurements that are obtained using this technique.

1.2 Research Objectives

The objective of this study is to apply and validate the HFTW technique for flux measurement. A secondary objective is to compare the HFTW technique with other flux measurement methods that are in use or development. To attain these objectives, we will attempt to find answers to the following questions:

1. How can the HFTW technique be implemented to measure flux?
2. How closely do HFTW flux measurements compare with actual values of mass flux?
3. What other techniques are currently available and in development to measure flux?
4. What are the relative costs, advantages, and limitations of each of the flux measurement techniques?

1.3 Research Approach

1. Based on the theoretical work presented in Goltz *et al.*, (2004), develop a

practical methodology for applying the HFTW technique to measure contaminant mass flux in the field under various conditions (regional groundwater flow velocity, orientation of HTFWs in relation to regional groundwater flow direction, HFTW pumping rates, etc.)

2. Apply the HFTW technique to measure a known mass flux under various conditions and compare values of known and measured flux. For this study, an ¡°artificial aquifer¡± will be used which will allow for the injection of a known flux o contaminant under controlled conditions.

3. Conduct a literature review of mass flux measurement methods and compare the costs, advantages, and limitations of these methods to the HFTW measurement technique.

1.4 Study limitations

- Validation of the HFTW method using an artificial aquifer is limited due to the fact that the aquifer does not truly represent conditions that will be encountered in the field. The artificial aquifer is homogeneous, well-controlled (constant boundary conditions, etc.), and on a relatively small scale in comparison to a natural system.

- While the HFTW method will be experimentally evaluated, the other innovative flux measurement methods that are included in this study (*e.g.* PFM and IGIM) will not be the subject of experiments. We will rely on literature reports to evaluate these other methods.

II. Literature review

2.1 Introduction

In this chapter, we review the literature regarding the four different methods that are currently used to measure contaminant mass flux. We begin with a discussion about the importance of being able to measure contaminant mass flux in order to address the problems of groundwater contamination described in chapter 1.

2.2 Background

As shown in chapter 1, the United States is facing a significant groundwater contamination problem. In order to comply with CERCLA and other environmental regulations at Department of Defense (DoD) installations, the DoD established the Defense Environmental Restoration Program (DERP, 2001). The DERP¡¯s 2001 Annual Report to Congress states that there are 28,500 contaminated sites requiring remediation throughout DoD (DERP, 2001). DoD has already spent approximately $25 billion in the last 17 years on restoration, and plans to spend $2 billion a year to remediate active and Base Realignment and Closure (BRAC) installations (DERP, 2001). The Air Force alone, in fiscal year 2001, obligated over $500 million to manage more than 6,000 contaminated sites at active and BRAC installations (DERP, 2001). Of these 6,000 sites, 1,462 are still under investigation and 700 sites have yet to be investigated (DERP, 2001). To manage a program of such magnitude and cost, prioritizing which sites receive funding is an important task, and prioritization decisions must be made based upon the best data (DERP, 2001). DoD ultimately plans to address all sites; however, due to limited resources, cleanup priority is placed on those sites posing the greatest risk to human health and to the environment (DERP, 2001).

Groundwater contamination by chlorinated solvents is particularly problematic, with contamination by chlorinated solvents found at approximately 80% of all Superfund sites with groundwater contamination (SERDP/ESTCP, 2001). Historically, pump-and-treat has typically been chosen as the strategy for managing contaminated groundwater. In fact, during the first few decades of the Superfund program, pump-and-treat was a component of the remedial remedy at 98% of over 600 Superfund sites with groundwater contamination. Unfortunately, especially at sites with chlorinated solvent source areas, pump-and-treat has proven to be incapable of achieving cleanup goals (SERDP/ESTCP, 2001).

During the last decade, due to the inability of conventional pump-and-treat technologies to achieve cleanup goals, scientists and engineers have investigated innovative plume management strategies, such as *in situ* biotic and abiotic technology applications, along with development of new approaches to remove or treat contaminant sources, such as *in situ* chemical oxidation, thermal technologies, and surfactant and cosolvent flushing (SERDP/ESTCP, 2001). In general, we can divide contaminant management strategies into two categories: (1) removal technologies and (2) containment technologies (Table 1).

Table 1. **Groundwater Remediation Strategies (API, 2003)**

Removal Technologies	Containment Technologies
Soil Vapor Extraction	Hydraulic Containment
Excavation	Barrier Walls / Cut-Off Trench
Air Sparging	Caps / Covers
Pump-and-Treat	Biological Barriers
LNAPL Skimming	
LNAPL Absorbents	
Total Combined Fluids Pumping	
Continuous Multi-Phase Extraction	
Bioslurping	
Natural Attenuation	

One of the most useful approaches to treat sites contaminated with organic contaminants is Monitored Natural Attenuation (MNA) (SERDP/ESTCP, 2001). The United States Environmental Protection Agency (USEPA, 1999) defines MNA as follows:

> The term "monitored natural attenuation" refers to the reliance on natural attenuation processes (within the context of a carefully controlled and monitored site cleanup approach) to achieve site-specific remediation objectives within a time frame that is reasonable compared to that offered by other more active methods. The "natural attenuation processes" that are at work in such a remediation approach include a variety of physical, chemical, or biological processes that, under favorable conditions, act without human intervention to reduce the mass, toxicity, mobility, volume, or concentration of contaminants in soil or groundwater. These in situ processes include biodegradation; dispersion; dilution; sorption; volatilization; radioactive decay; and chemical or biological stabilization, transformation, or destruction of contaminants.

MNA can not only be an economical alternative by itself to manage a large plume, but it can also be used effectively in conjunction with other remediation technologies.

In order to assess the protectiveness of natural attenuation, as well as to evaluate the efficacy of engineered remediation systems, groundwater models are important tools. Models can be used both to predict how the distribution of contamination in space and time is affected by natural and engineered processes, and to help design remediation technologies. A crucial component of a groundwater contaminant fate and transport model is the contaminant source term. Source terms are normally incorporated into models as either contaminant concentration boundary conditions or contaminant flux boundary conditions. In order to develop contaminant fate and transport models that reflect actual site conditions and processes, it is necessary to have relatively accurate concentration and flux measurements to use in the model as boundary conditions.

As is discussed in some detail below, the ability to measure mass flux of a groundwater contaminant is important so that we may be able to assess the relative risk posed by a contaminated site, evaluate remediation technologies that are being developed and tested, evaluate the efficacy of MNA at a site, and model the transport and fate of contaminants in the subsurface.

2.3 Need for flux measurement

2.3.1 Prioritization of cleanup

A contaminant source zone may have the majority of contaminant mass located within low permeability regions. In this case, even though contaminant mass and dissolved concentration may be large, the flux of contaminant leaving the source zone will be relatively low. Conversely, a smaller source zone in a high permeability region may result in significant contaminant mass flux leaving the area. With this in mind, Einarson and Mackay (2001) contend that to assess the risk to receptors of groundwater

contamination, contaminant mass flux, rather than contaminant concentration, should be evaluated.

In their paper, Einarson and Mackay (2001) demonstrate how knowledge of the contaminant mass flux emanating from a contaminant source area can be used to estimate the contaminant concentration at a downgradient water supply well. After making a number of simplifying assumptions, Einarson and Mackay (2001) show that the contaminant concentration (C_{sw}) in a downgradient water supply well pumping at rate Q_{sw} can be calculated as:

$$C_{sw} = M_f \times A \div Q_{sw} \qquad (1)$$

where M_f is the contaminant mass flux $[ML^{-2}T^{1}]$ emanating from a contaminant source area whose plume is captured by the supply well and A $[L^2]$ is the area of the plume orthogonal to the groundwater flow direction that is captured by the well.

To demonstrate how the measurement of contaminant flux from a source zone is related to risk, and therefore, useful in prioritizing site cleanups, suppose there are two different contaminated sites that have a source zone and supply well at each site (Figure 5) (Einarson and Mackay, 2001).

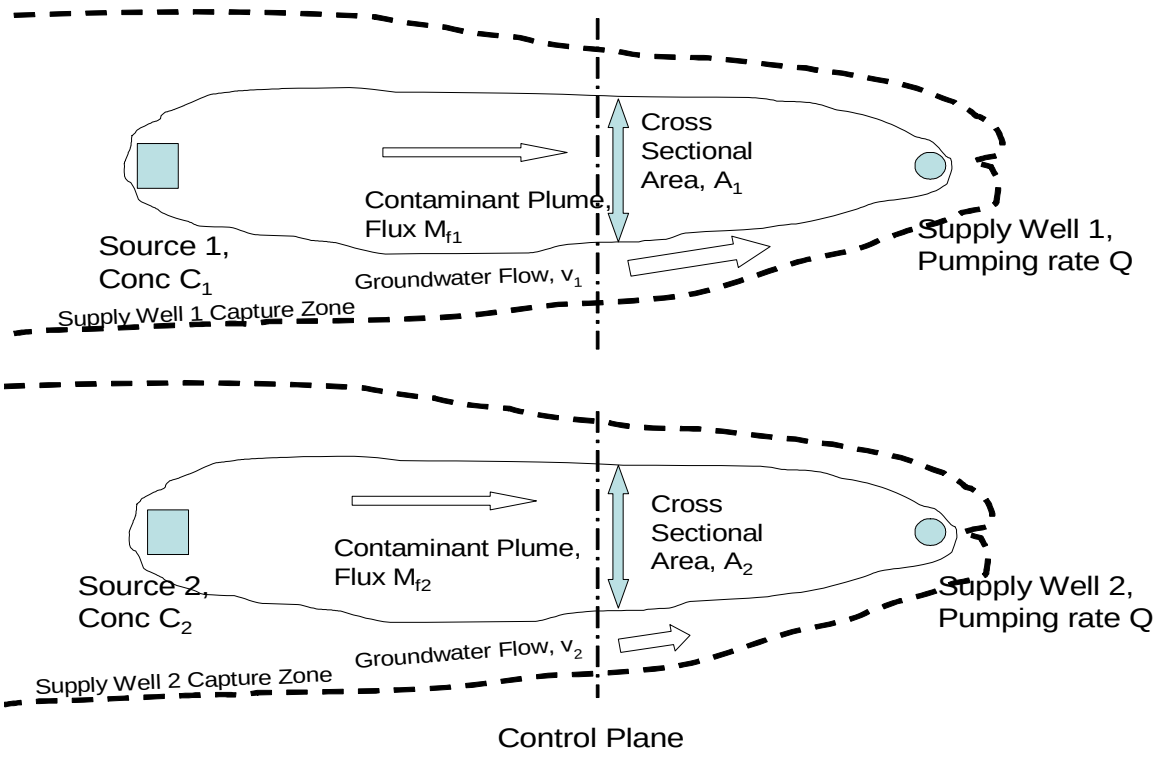


Figure 5. Plan view of two hypothetical contaminated sites (Einarson and Mackay, 2001).

Just downgradient of Source 1, dissolved concentrations of contaminant are measured at 1 *mg/L*, while just downgradient of Source 2, contaminant concentrations are 5 *mg/L*. Let us assume the cross-sectional areas of the two plumes are the same at the control planes shown in Figure 5 *($A_1 = A_2$)* and that the groundwater velocities measured at the control planes are 2 *m/d* and 0.1 *m/d* for Sources 1 and 2, respectively. Measurements of contaminant flux downgradient of the two sources indicate that the flux from Source 1 is 2 *g/(m^2-d)*, while the flux leaving Source 2 is 0.5 *g/(m^2-d)*. The plume from each source is captured by a supply well that is pumping at a constant rate *Q*. In this hypothetical case, even though Source 2 has a higher downgradient contaminant

concentration, application of Equation (1) shows that Source 1 will result in a higher concentration in Supply Well 1 than the concentration seen in Supply Well 2 resulting from Source 2. This, of course, is due to the greater mass flux leaving Source 1. Thus, when prioritizing the two sites for cleanup, a decision maker might decide to address remediation of Site 1 first, even though Site 2 has higher contaminant concentrations.

As described above, it is contaminant mass flux, rather than contaminant concentration, that is more crucial in determining the risk posed by a contaminant source and plume. Thus, ideally, site managers and regulators will have access to accurate flux measurements in order to inform their site management decisions.

2.3.2 Evaluating the efficacy of cleanup technologies

The Strategic Environmental Research and Development Program (SERDP) and the Environmental Security Technology Certification Program (ESTCP) (SERDP/ESTCP, 2001) reported that assessing the effects of source zone treatment is one of the highest priorities needs for science and technology within the remediation area. As we attempt to evaluate the various source remediation technologies that are being proposed and fielded, we must keep in mind that the measure of technology success is risk reduction (as opposed to mass reduction, concentration reduction, or some other measure). As demonstrated in the section above, flux reduction can be directly tied to risk reduction, so being able to measure reduction of flux by comparing pre- and post-remediation fluxes, is crucial to being able to evaluate the efficacy of source zone remediation technologies (SERDP/ESTCP, 2001; Soga *et al.*, 2002).

A number of recent studies have been concerned with how application of source remediation technologies may result in flux reduction (Sale and McWhorter, 2001; Rao *et*

al., 2001; Soga *et al.,* 2002; Rao and Jawitz, 2003; McWhorter and Sale, 2003; NAS, 2004; Lemke *et al.,* 2004). Soga *et al.* (2002) focused upon how flux reduction may be a function of the interactions between the remediation technology, source morphology, and subsurface heterogeneities. Some technologies can increase or decrease the long-term contaminant flux in downgradient receptor areas by changing the source morphology during treatment, while other technologies can not change the mass flux because they treat only the plumes without touching source areas (Soga *et al.,* 2002).

Rao *et al.* (2001) conducted three-dimensional particle-tracking model simulations for heterogeneous flow fields and field experiments at the Dover AFB, Delaware to show that significant contaminant flux reductions can be achieved by partial removal of contaminant mass from DNAPL source zones. Furthermore, Rao and Jawitz (2003) used a stream tube model to theoretically calculate how reduction of contaminant mass flux is related to reduction of source mass for homogeneous and heterogeneous media. Assuming a homogeneous distribution of DNAPL, and quantifying hydraulic conductivity heterogeneity using the standard deviation of the groundwater velocity distribution (¥ò),Rao and Jawitz (2003) showed that for increasingly heterogeneous media, relatively small source mass reductions could lead to relatively significant flux reductions (Figure 6).

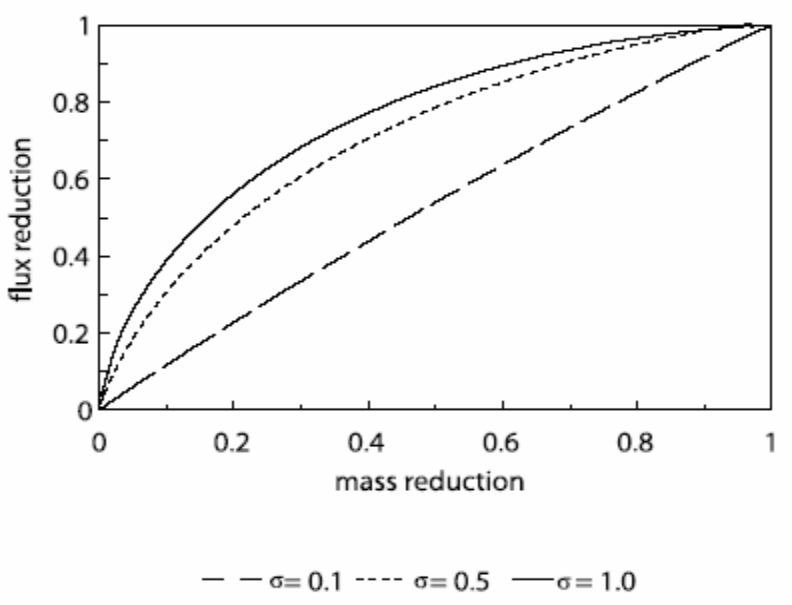


Figure 6. Fractional reductions in contaminant flux as a function of source mass removal for three values of the standard deviation (¥ò) of the groundwater velocity distribution (Rao and Jawitz, 2003)

Rao and Jawitz (2003) explained this based on the key assumption that DNAPL remediation technologies will preferentially remove or destroy DNAPL in high hydraulic conductivity zones (represented by high velocity stream tubes). Thus, removal of the relatively small fraction of the total DNAPL mass that resides in the high velocity stream tubes can result in relatively large flux reductions, as it is this fraction that contributes the most to mass flux leaving the source area. Even though significant contaminant flux reductions are realized through partial mass reduction in the DNAPL source zone, it is still a matter of debate whether such mass flux reduction is sufficient to achieve adequate risk reduction and regulatory compliance (Rao *and* Jawitz, 2003).

The National Academy of Sciences (NAS, 2004) also showed that mass removal may result in a substantial reduction in mass flux (Figure 7). In agreement with the study by Rao and Jawitz (2003), the NAS (2004) suggests that for a given reduction in

mass, mass flux reduction in a heterogeneous aquifer may be significantly greater than for a homogeneous formation (Figure 7). Lemke *et al.* (2004) also used modeling to predict that removal of 60 to 99% of contaminant source mass can reduce mass flux under natural gradient conditions by approximately two orders of magnitude.

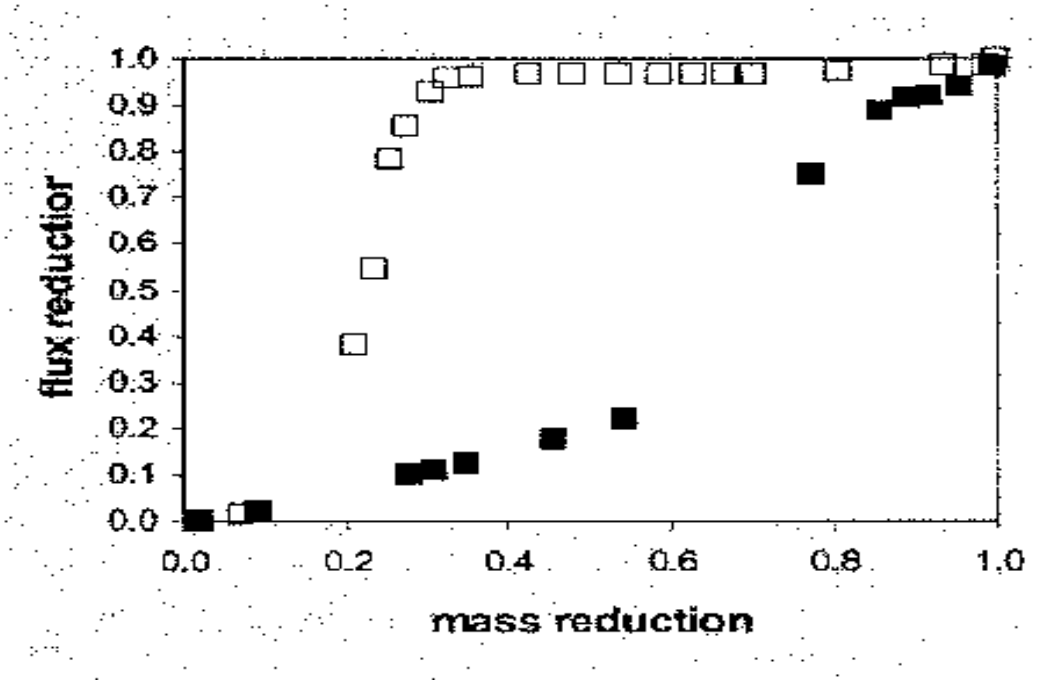


Figure 7. Simulated contaminant flux reduction as a function of mass reduction; open squares represent heterogeneous sites and solid squares represent homogeneous sites (NAS, 2004)

In contrast to the results discussed above (*e.g.* Rao and Jawitz, 2003), Sale and McWhorter (2001) used an analytical model with a homogeneous flow field and heterogeneous DNAPL distribution to show that significant flux reductions could only be achieved if there were significant reductions in contaminant mass. This result corresponds to the homogeneous site simulation depicted in Figure 7 (NAS, 2004). McWhorter and Sale (2003) argued that the conclusion that significant flux reduction

could be achieved with relatively low mass removal was incorrect because of faulty assumptions employed by Rao and Jawitz (2003). Specifically, Rao and Jawitz (2003) assumed: (1) complete depletion of DNAPL within individual stream tubes and (2) no mass transfer between stream tubes. These assumptions can create positive bias in terms of benefits that can be achieved from partial depletion of DNAPL mass. Thus McWhorter and Sale (2003) insisted that even though the potential benefits of partial mass reduction may include reduced risk, reduced source longevity, reduced site-care requirements, and enhanced natural attenuation, quantification of such benefits as a function of mass removal is necessary. Clearly, the ability to accurately measure contaminant flux is crucial to quantifying the benefits of applying a source remediation technology.

2.3.3 Quantifying natural attenuation (NA).

Natural attenuation is an important strategy that is used to manage groundwater contamination (SERDP/ESTCP, 2001). A number of studies have measured contaminant flux or mass discharge in order to quantify the extent of NA (Borden *et al.,* 1997; Bockelmann *et al.,* 2003; Peter *et al.,* 2004).

If one assumes that physical attenuation processes (*e.g.* dispersion, volatilization, sorption) are steady or small, measurements of mass flux through control planes located perpendicular to the principal contaminant flow direction at different distances from the contaminant source can be used, along with the average travel time between the control planes, to estimate an effective first-order contaminant decay coefficient (Borden *et al.,* 1997). The assumptions of steady-state flow, dispersion, and sorption appear reasonable at many contaminated sites (Bockelmann *et al.,* 2003) and a number of studies have

demonstrated that volatilization of organic contaminants is not significant. For example, McAllister and Chiang (1994) showed that volatilization accounted for only 5 to 10% of the mass reduction of volatile BTEX compounds. With these assumptions of steady or insignificant physical attenuation processes, measured flux reductions may be regarded as primarily resulting from chemical or biological degradation of the contaminant (Bockelmann *et al.*, 2003).

The rate of NA at a site depends on the site¡¯s unique geochemical character. Borden *et al.* (1997) used mass flux measurements to demonstrate methyl tert-butyl ether (MTBE) and BTEX natural attenuation in a shallow aquifer contaminated by leaking underground storage tanks (USTs) that contained gasoline and diesel fuel. In the study, flux measurements were used to show that NA was higher near the source area than downgradient and that NA of the BTEX compounds was generally greater than NA of MTBE (Borden *et al.*, 1997). Studies such as these show that mass flux measurement is a powerful tool that can be used to evaluate NA at contaminated field sites, thus provide decision makers with important information that they can use to manage risk.

2.3.4 Modeling fate and transport (source term to determine downgradient concentration)

Groundwater modeling has developed tremendously over the past 25 years, and we now have the ability to quantitatively estimate groundwater flow and contaminant mass transport in the subsurface (Bedient *et al.*, 1994). The purposes of modeling are as follows (Bedient *et al.*, 1994):

1. Testing a hypothesis, or improving knowledge of a given aquifer system.
2. Understanding physical, chemical, or biological processes.

3. Designing remediation systems.
4. Predicting future conditions or the impact of a proposed stress on a ground water system.
5. Resource management.

After developing a conceptual model of a system, it is necessary to translate the conceptual model into a mathematical model consisting of governing equations and initial and boundary conditions in order that the value of the dependent variable of interest (*e.g.* contaminant concentration) can be determined as a function of space and time (Wiedemeier *et al.*, 1999). For fate and transport modeling, boundary conditions are specified in terms of contaminant concentrations and/or fluxes (Wiedemeier *et al.*, 1999). It is apparent that being able to measure contaminant flux is critical to our ability to model contaminant fate and transport at a site, and hence, our ability to use models to support management decisions at the site.

2.4 Flux measurement methods

2.4.1 Transect method

The conventional method for measuring contaminant mass flux in a plume is to install transects of monitoring wells along control planes that are orthogonal to the direction of groundwater flow (See Figure 2). Either single-screen or multilevel groundwater monitoring wells can be used for this purpose (API, 2003). Groundwater samples are collected at various points in the control planes, and contaminant concentrations measured at these points. Note that, in order to determine total contaminant mass discharge through the control planes, it is necessary that the monitoring wells sample the entire width and depth of the plume.

Applying the transect method to determine mass flux and discharge is

straightforward. After having measured the contaminant concentration (C_i) at the i^{th} sampling point, the advective mass flux, $M_{f,i}\left[M/L^2T\right]$, at the point can be calculated as:

$$M_{f,i} = C_i \times q_i \qquad (2)$$

where $q_i\left[L/T\right]$ is the groundwater specific discharge at well *i* (Bockelmann *et al.*, 2003). The groundwater specific discharge is defined by Darcy¡¯s Law as the product of the hydraulic conductivity at well *i* (K_i) and the hydraulic gradient (∇h) ($q = -K_i \nabla h$). We can determine the hydraulic gradient from a potentiometric surface contour map that is constructed based on static water level measurements at the monitoring points. Hydraulic conductivity can be obtained using appropriate slug test or pumping test methods (Weight and Sonderegger, 2001).

The contaminant mass discharge for individual sampling points, $M_{d,i}\left[M/T\right]$, and the total mass discharge through the control plane, $M_d\left[M/T\right]$, are defined as:

$$M_{d,i} = C_i \times q_i \times A_i = M_{f,i} \times A_i \qquad (3)$$

$$M_d = \sum_{i=1}^{n} M_{d,i} \qquad (4)$$

where *n* is the number of monitoring points in the control plane and $A_i\left[L^2\right]$ represents the area of the control plane associated with the i^{th} monitoring point. This area may be estimated by constructing Theissen polygons (polygons whose sides are perpendicular bisectors of lines connecting adjacent monitoring points) in the control planes (Borden *et al.*, 1997; Bockelmann *et al.*, 2003). The average mass flux (M_f) can be obtained by dividing the total mass discharge by the cross-sectional area of the plume at the control

plane (A):

$$M_f = M_d / A \quad (5)$$

By combining equations (3) and (4), we also see that average mass flux can be directly calculated from the mass flux measurement at each sampling point as follows:

$$M_f = \frac{\sum_{i=1}^{n} M_{f,i} A_i}{A} \quad (6)$$

The limitation of the transect method is a result of the fact that sampling is at discrete points across the direction of flow, so a large representative volume of the subsurface is not necessarily interrogated. Increasing the detail of sampling, in order to account for spatial heterogeneities, or the range of sampling, to encompass the entire plume cross-section, requires increasing the number (and therefore cost) of sampling wells (Bockelmann *et al.*, 2003). Guilbeault *et al.* (2005) showed that even for a relatively homogeneous aquifer, vertical well spacing as small as 15 *cm* and lateral spacings between 1 and 3 *m* are needed to characterize small zones of high concentration near a NAPL source.

Borden *et al.* (1997) evaluated the mass flux of dissolved gasoline constituents (BTEX and MTBE) released from an underground storage tank using this transect method in a Coastal Plain aquifer in rural Sampson County, North Carolina in 1997. Using mass discharge measurements at four control planes, the authors estimated the field scale first-order natural attenuation decay rate of the dissolved contaminants. One advantage of this mass discharge approach to evaluating the rate of natural attenuation is that it does not require fitting a solute transport model to concentrations at individual wells in order to obtain a degradation rate constant. A disadvantage of the approach is

that since it is based on sampling at discrete points, the sparser the points, the less reliable the mass discharge estimate compared to estimates based on volume-averaged approaches (such as the IGIM and HFTW methods) which will be discussed below (Bockelmann *et al.*, 2003).

2.4.2 Passive flux meter (PFM)

This newly-developed method is a point method (in that sense, similar to the transect method), that involves placing PFMs at points along a control plane to intercept contaminated groundwater. The PFM consists of permeable sorbents and resident tracers (Hatfield *et al.*, 2001; Hatfield *et al.*, 2004; Jonge and Rothenberg, 2005). Hydrophobic and hydrophilic permeable sorbents retain dissolved organic and/or inorganic contaminants that are present in the fluid that passes through the PFM. These sorbents have 3resident tracersFwhich leach into the groundwater at rates proportional to fluid flux. The cumulative volume of groundwater that passes through the flux meter can be calculated using an analytical model that accounts for the mass of resident tracer that has desorbed into the water. Knowing the cumulative volume of groundwater that has passed through the PFM, as well as the time the PFM has been in place and the effective cross-sectional area of the PFM screens, specific discharge of the groundwater can be calculated (Hatfield *et al.*, 2001). The contaminant mass retained in the flux meter sorbent over the time the PFM has been in place can be used, in combination with the groundwater flux, to determine the contaminant mass flux at the PFM. As this is a point method, the flux measured at each PFM can be summed, using the methods described in Section 2.4.1 (see equations (3) through (6)), to obtain an average flux and a total mass discharge over the plume cross-section.

One advantage of this method over the transect method is that the flux measured by the PFMs is averaged over the time the PFM is in place. This is particularly relevant when discharge varies significantly with time. This temporal averaging may help circumvent overestimation or underestimation of flux that may result from a point measurement in time. Another advantage of the PFM method is that groundwater specific discharge is measured directly. This is in contrast to the transect method, which requires separate measurements of hydraulic conductivity and groundwater gradient in order to apply Darcy's law to determine specific discharge. As with the transect method, properly installed PFMs should intercept the entire width and depth of a plume of dissolved contaminant.

As a point method, the flux meter method has the same disadvantages as the transect method. That is, increasing the detail of sampling, in order to account for spatial heterogeneities, or the range of sampling, to encompass the entire plume cross-section, requires increasing the number (and therefore cost) of installed PFMs.

In a laboratory column experiment, Campbell *et al.* (2004) demonstrated this method as a promising technique for determination of specific discharge and contaminant flux. In the experiment, the PFM measured values for specific discharge and chrome (*VI*) mass flux that were within 19% and 17% of the actual discharge and flux values, respectively.

Hatfield *et al.* (2001) used the PFM technique with four flux meters to measure specific discharge in an artificial box aquifer (52 *cm* long by 30 *cm* high and 37 *cm* deep) within 2.5 percent of the true discharge. The investigators also used the PFM technique to estimate contaminant mass flux within 6.8% of the true flux. Hatfield *et al.* (2004)

measured 2,4-Dimethyl-3-pentanol (DMP) fluxes using multiple PFMs in an artificial box aquifer (27 *cm* by 20 *cm* by18 *cm*). DMP flux measurements were all within 5 % of their actual values. The investigators found that the accuracy of the mass flux measurement generally increases with the total volume of water intercepted by the PFM. That is, the longer the PFM is in place or the greater the natural groundwater flow rate, the more accurate the flux measurement.

Jonge and Rothenberg (2005) demonstrated the PFM technique in long-term laboratory experiments, using unsaturated soil columns (20 *cm* by 20 *cm*). The investigators found that if the correct adsorbent was used in the PFM, flux of phenanthrene and glyphosate could be measured with an accuracy of 3.6% ~ 17.8% and 12.4% respectively.

2.4.3 Integral groundwater investigation method (IGIM)

Spatially integrated contaminant mass discharge (M_d) can be estimated by pumping potentially contaminated water at one or more wells located along a control plane downgradient of a suspected pollutant source zone so as to fully capture the contaminant plume emanating from the source (Figure 8) (Bockelmann *et al.*, 2003; Bauer *et al.*, 2004). The number and location of the wells, along with pumping rates and times, must be chosen to ensure that the entire plume is captured, in order to determine the total mass discharge across the control plane.

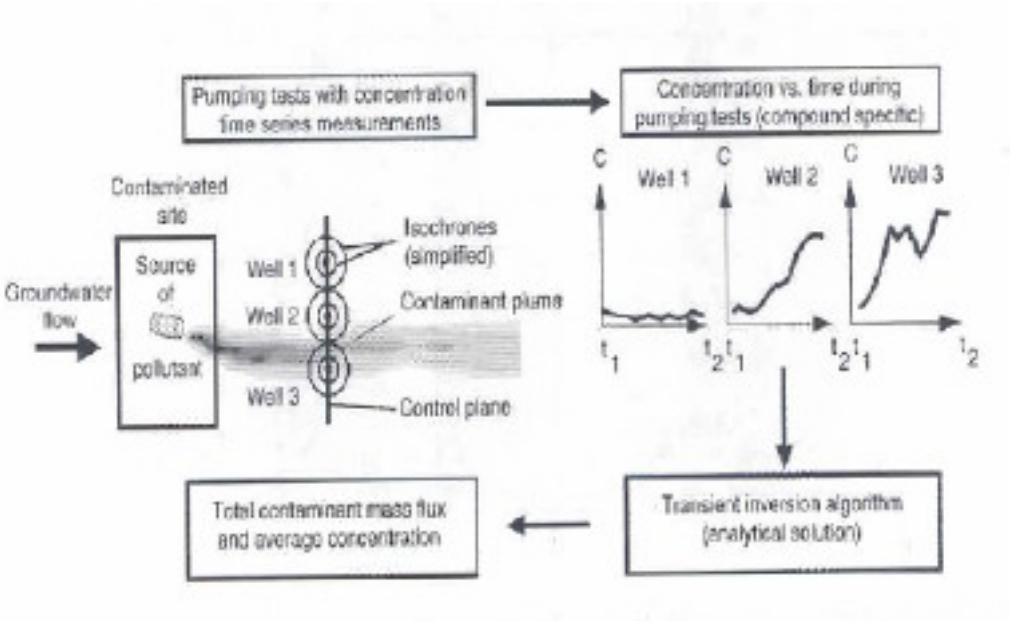


Figure 8. Application of integral groundwater investigation method (Bockelmann *et al.*, 2001)

Mass discharge is determined by monitoring contaminant concentration at each of the pumping wells vs. time (Figure 8). Under the following assumptions: (1) the flow towards the abstraction wells is radially symmetrical, *i.e.* the natural flow can be neglected during the pumping test; (2) the aquifer is homogeneous with regard to porosity, hydraulic conductivity and thickness, and (3) the concentration does not vary significantly along each of the streamtubes at the scale of the well capture zone, although it may vary from streamtube to streamtube, Bockelmann *et al.* (2003) described and applied a method at a contaminated site to analytically invert the concentration versus time (*CT*) measurements to obtain an estimate of mass discharge across a control plane perpendicular to the direction of groundwater flow. Bockelmann *et al.* (2003) also noted that for a heterogeneous aquifer, where there are detailed measurements of the hydraulic conductivity distribution in space, the *CT* data can be numerically inverted to estimate

mass discharge. If we are able to quantify the cross-sectional area of the plume captured by the extraction wells, the average mass flux, $M_f \left[M / L^2 T \right]$, can be obtained by dividing the total mass discharge by the cross-sectional area.

Because the IGIM is based on pumping wells, the method can interrogate a large volume of the subsurface with installation of relatively few wells as compared to point methods. The associated disadvantage of this is that extraction of potentially contaminated water can result in safety concerns and water treatment/disposal costs (Bockelmann *et al.,* 2003). Since it is a pumping technique, the IGIM will not work in geologies with low transmissivities. The method also requires capture of the entire plume--incomplete capture will result in underestimation of the mass discharge. On the other hand, if the IGIM well capture zone is too large, contaminant from the plume may mix with large volumes of uncontaminated water, resulting in *CT* responses at the wells where the concentrations are below analytical detection limits. Asymmetrical well capture zones around a well caused by significant heterogeneities lead to uncertain control plane width. Also, preferential flowpaths across the control plane could be overestimated or underestimated by using the average groundwater flux at the scale of the individual well capture zone (Bockelmann *et al.,* 2003).

Bockelmann *et al.* (2001; 2003) and Peter *et al.* (2004) applied the IGIM to estimate the NA of a petroleum hydrocarbon contaminant plume at a former gasworks site in Southwest Germany. Bockelmann *et al.* (2003) quantified mass fluxes and NA rates using the transect and IGIM methods at two control planes. The investigators showed that due to the dependence of the transect method on concentration measurements at points in a relatively sparse monitoring network, there was considerable

uncertainty in the flux measurement. Considerable differences (97% ~ 159%) were noted between the fluxes measured by the two methods at the two control planes (Bockelmann *et al.*, 2003). The investigators attributed the differences in the two methods to the fact that the transect method was inadequate in capturing the plume and geologic heterogeneities and concluded that the IGIM was a viable method for mass flux measurement.

The study by Bockelmann *et al.* (2003) also quantified NA rate constants using both the IGIM and W*enterline*[3] point scale approaches. The centerline approach made use of a long-term tracer test to delineate the contaminant transport path and compare contaminant concentration reduction with the concentration reduction of a conservative tracer along the plume centerline. Both approaches resulted in similar NA rate constant values.

Bauer *et al.* (2004) quantified PCE and TCE mass fluxes by using both a numerical inversion code, CSTREAM (Bayer-Raich *et al.*, 2003), and a simplified analytical approach to interpret IGIM data from an industrialized urban area in Linz, Austria. The results of the numerical and analytical approaches deviated by less than a factor of two.

The IGIM was also evaluated as a component of the European Union-sponsored Integrated Concept for Groundwater Remediation (INCORE, 2003) project at four European cities. The INCORE (2003) studies involved quantification of chlorinated hydrocarbon contaminant flux at four sites. From the INCORE (2003) studies, the investigators concluded that the IGIM was capable of quickly and with certainty estimating the average contaminant concentration, spatial distribution of concentration

values along a control plane, and discharge downgradient of a contamination source zone.

2.4.4 Horizontal Flow Treatment Wells (HFTWs)

HFTWs consist of two pumping wells, with each well having extraction and injection screens, in order to circulate contaminated water in the subsurface without the need to extract it aboveground (See Figure 4). In an HFTW well-pair, one well pumps water upwards while the other pumps downwards. Operation of these wells results in a capture zone upstream of the wells, as well as a recirculation zone between the wells (Figure 9) (Christ *et al.*, 1999).

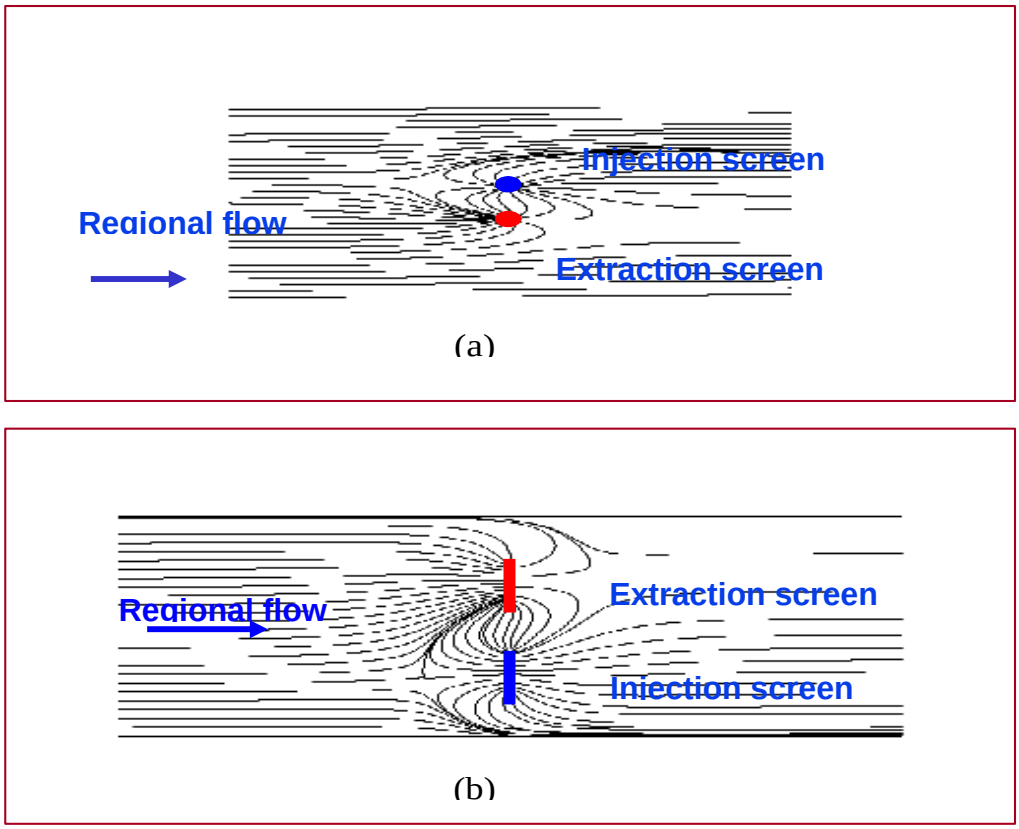


Figure 9. **(a) Plan view in upper horizon of an aquifer and (b) cross sectional view at the down flow well depicting HFTW operation (after McCarty *et al.*, 1998)**

The plan view of Figure 9 depicts the streamlines in the upper horizon of the aquifer, where the upflow well has an injection screen and the down flow well has an extraction screen, and the cross sectional view shows the stream lines at the injection and extraction screens of the downflow well. McCarty *et al.* (1998) applied HFTWs in the field for contaminant plume cleanup and the flow field that results from operation of these wells has been analytically modeled by Christ *et al.* (1999) and numerically modeled by Gandhi *et al.* (2002).

Goltz *et al.* (2004) and Huang *et al.* (2004) proposed an innovative approach to measure flux by operating HFTWs. We have seen that mass flux can be determined by measuring contaminant concentration, and aquifer hydraulic gradient and conductivity (Equation (2)). With the pumps in the HFTWs turned off, hydraulic gradient may be determined by measuring the piezometric surface at the two HFTWs and a third piezometer. Volume-averaged contaminant concentration in the HFTWs can be measured as contaminated water flows through the wells. With these two parameters measured relatively straightforwardly, we see the crucial parameter in determining mass flux is the hydraulic conductivity.

Goltz *et al.* (2004) proposed and tested two basic approaches for using HFTWs to measure hydraulic conductivity. The first approach was based on the dipole flow test method (Kabala, 1993) while the second approach relies on a tracer test to measure interflow between the two HFTWs.

The multi-dipole method extends the dipole method by applying it to obtain an estimate of hydraulic conductivity during operation of an HFTW system (Goltz *et al.*, 2004). A dipole is a dual-screen well; in essence, it is the upflow well of an HFTW

well-pair. Assuming homogeneity, steady-state flow, and superposition, Goltz *et al.* (2004) developed an analytical solution relating the drawdown and mounding measured at the downflow and upflow HFTWs, respectively, to horizontal and vertical hydraulic conductivity. As the equation is nonlinear, Goltz *et al.* (2004) also presented a method that made use of a genetic algorithm to determine the values of horizontal and vertical conductivity that best fit the drawdown/mounding data obtained from operating the HFTW system at several flow rates.

The interflow measurement approach uses a tracer test to measure interflow of water between the HFTWs, where interflow is defined as the fraction of water flowing into an extraction well screen that originated in one of the two injection screens. The test consists of injecting a step concentration of a tracer into the upflow well and a step concentration of a second tracer into the downflow well. Subsequently, tracer concentrations at each of the four screens of the HFTW well-pair are measured. Assuming steady-state, mass balance may be used to formulate four equations with four unknowns, where the unknowns are the interflows of water between the four injection-extraction well screen pairs. Solving for these measured interflows, a three-dimensional flow model, MODFLOW (Harbaugh and McDonald, 1996), is used in conjunction with a genetic algorithm to obtain values of horizontal and vertical conductivity that result in the best fit of the HFTW flow model in MODFLOW to the measured interflow data (Goltz *et al.*, 2004).

The HFTW flux measurement method has the benefit of the volume-averaged IGIM, in that rather than measuring flux at points, the method, through pumping, interrogates a large volume of the subsurface. It achieves this benefit while avoiding the

costs of extracting contaminated groundwater from the subsurface.

Goltz *et al.* (2004) conducted an experiment in an artificial aquifer located in Canterbury, New Zealand, to measure hydraulic conductivities using various measurement techniques. Assuming isotropy, which was reasonable in the case of the relatively homogeneous sand aquifer, the investigators used the HFTW interflow approach described above to determine a hydraulic conductivity of 0.16 *cm/sec*. This compared well with the ¡°actual¡± conductivity of the aquif of 0.17 *cm/sec*, which was measured previously in a number of tests (Bright *et al.*, 2002). When horizontal and vertical conductivities were not constrained to be equal, underestimated values of 0.13 and 0.094 *cm/sec* were obtained for k_r and k_z respectively. Apparently, assuming anisotropy for an aquifer that is relatively isotropic leads to a significant underestimate of the conductivity when using the HFTW interflow approach.

A preliminary test of the technique to measure the flux of a conservative tracer in the artificial aquifer was also accomplished by Huang *et al.* (2004). In that test, the measured mass flux of a tracer was within 23% of the actual value.

III. Methodology

3.1 Introduction

Detailed procedures for measuring mass flux using the HFTW and conventional transect methods are described in this chapter. In section 3.2, the artificial aquifer which will be used for the flux measurement experiments is described. In section 3.3, installation and operation of the HFTWs in the artificial aquifer is explained. In Section 3.4 we provide details on the two approaches we will take to measure hydraulic conductivities and mass fluxes with the HFTWs; the multi-dipole approach and the tracer test approach. In section 3.5, we describe the process of mass flux measurement using the conventional transect method. Finally, our methodology for costing each of the mass flux measurement methods is laid out in section 3.6.

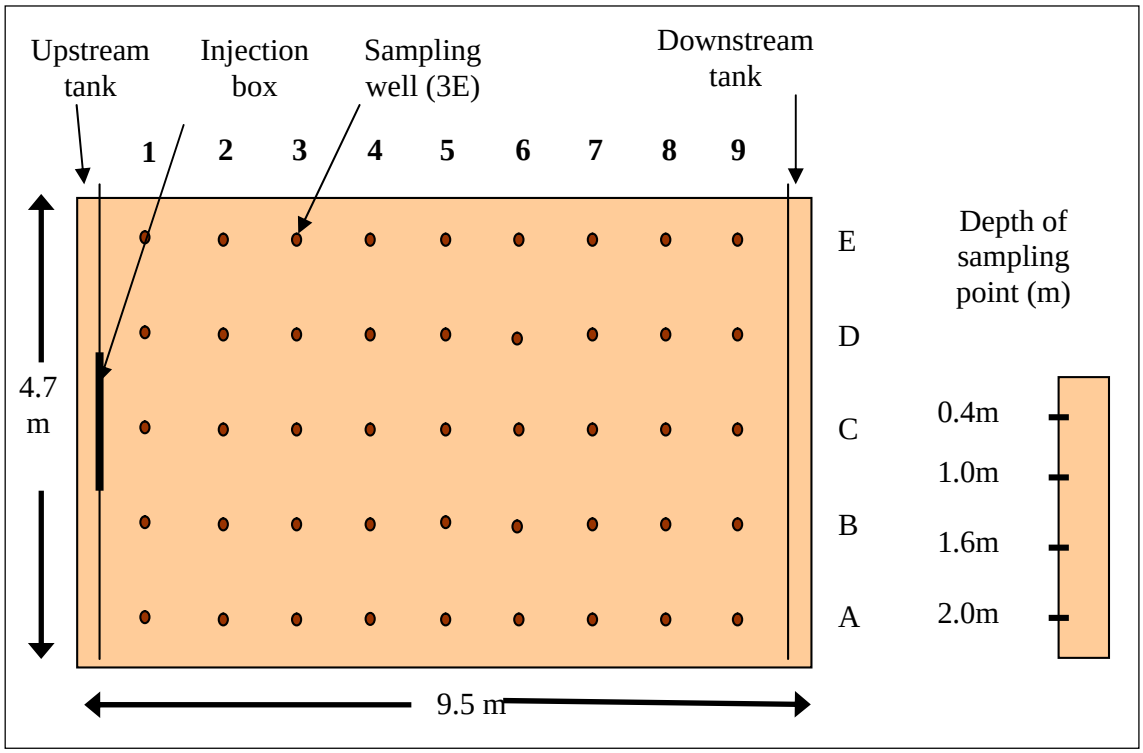
3.2 Artificial aquifer

Before conducting a full-scale field experiment to evaluate the HFTW flux measurement method, a ¡°mes-scale¡± evaluation in anartificial aquifer has been proposed (Goltz, 2004). Such a meso-scale evaluation is an intermediate step between well-controlled laboratory studies (typically conducted in one- or two-dimensions) and expensive, largely uncontrolled field studies. The proposed evaluation of the HFTW and transect mass flux measurement techniques will be conducted in a large three-dimensional, confined artificial aquifer in Canterbury, New Zealand, which was used for the contaminant transport experiment described by Bright *et al.* (2002) (Figure 10).

The inner dimension of the homogeneous sand aquifer is 9.5 *m* long, 4.7 *m* wide, and 2.6 *m* deep. The aquifer is filled with coarse sand that was dry sieved to fall within the size range 0.6 to 1.2 *mm* in diameter. Constant-head tanks (0.75 *m* long, 4.7 *m* wide

and 3.1 *m* high) that control the hydraulic gradient of the aquifer, bound the aquifer¡¯ upstream and downstream ends. The aquifer is operated under confined conditions, with the top surface sealed with a plastic liner. The bottom and sides of the aquifer are no-flow boundaries lined with impermeable butyl rubber.

(a)



(b)

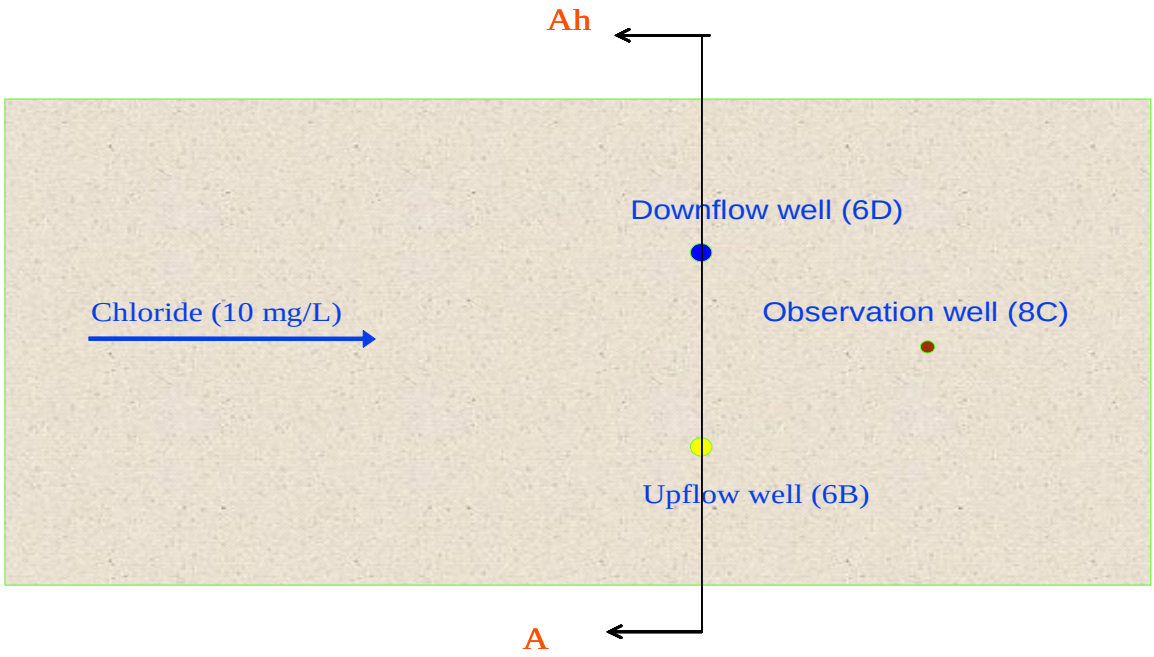
Figure 10. **(a) Artificial aquifer used in the HFTW experiment, Canterbury, New Zealand (b) Plan view of sampling well distribution in the aquifer and the vertical distribution of sampling points in a sampling well (Bright *et al.*, 2002)**

As shown in Figure 10, there are 45 wells installed on a 1 *m* by 1 *m* grid, with 9 rows down and 5 rows across the aquifer. The first row of wells is located 0.75 *m* down-gradient from the header tank and the last row is located 0.75 *m* up-gradient from the end tank. The middle row of wells is located down the center of the aquifer, with the outer rows of wells located about 0.35 *m* from each side wall. Each well is a 2.5 *cm* diameter tube extending to the bottom of the aquifer. The wells are slotted throughout their length and covered with a nylon sock to prevent entry of sand. As shown in the figure, most of the wells have four sampling ports at depths of 0.4 *m*, 1.0 *m*, 1.6 *m*, and 2.2 *m* below the top of the aquifer, with two wells having seven sampling points. Each sampling port consists of a 7.5 *cm* long section of well screen with a Teflon sample tube extending from the sampling depth to an automatic sample collector. Computer controlled peristaltic pumps enable fully automated water sampling from the 180 sampling points (Bright *et al.*, 2002, Goltz *et al.*, 2004).

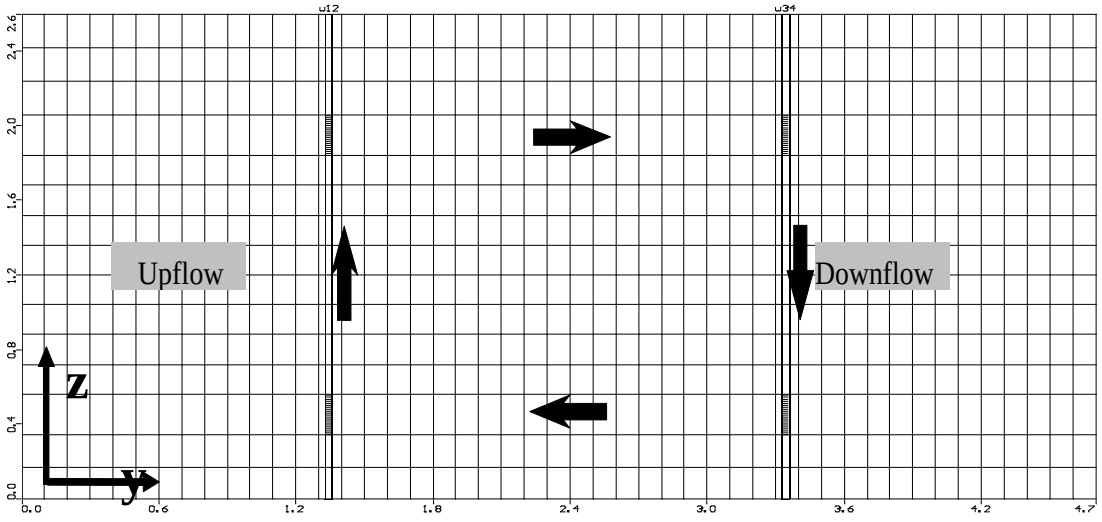
3.3 HFTW installation and operation

3.3.1 The process of HFTW installation and operation

An HFTW well pair along with a single observation well was installed in the artificial aquifer at locations 6B, 6D, and 8C (the upflow HFTW at 6B, the downflow at 6D, and the observation well at 8C) as shown in Figure 11 (Goltz *et al.*, 2004).



Plan view



Cross-section A-A

Figure 11. **Plan and cross-section views showing two HFTWs and observation well (Goltz *et al.*, 2004)**

The injection screens (the upper screen of the upflow well and the lower screen of the downflow well) and the extraction screens (the lower screen of the upflow well and

the upper screen of the downflow well) are constructed using 2.5 *cm* diameter PVC. The injection/extraction screens are 22.5 *cm* long, each consisting of two 7.5 *cm* long PVC slotted sections separated by a 7.5 *cm* long PVC blank. The injection and extraction screens in each well are separated by 1.28 *m*, with the upper and lower end of each screen isolated using inflatable rubber packers. Two pumps are used (one for each HFTW) to extract water from the extraction screen and inject water into the injection screen at a specified flow rate.

Water containing chloride as a model contaminant will be continuously input at the header tank. After measuring the water levels in the observation well at location 8C and at the two HFTWs to calculate the magnitude and direction of the hydraulic gradient, the HFTW pumps will be turned on. Bromide and nitrate tracers are injected into the injection screens of the upflow and downflow wells, respectively. Injection of tracers will be continued until steady-state bromide and nitrate concentrations are reached at the two extraction screens. Concentrations of bromide, chloride, and nitrate will be measured over time at all four HFTW screens, for application of the tracer approach. During operation of the HFTWs, steady-state drawdown at the downflow well and mounding at the upflow well will be measured for application of the multi-dipole approach. The above-described experiment will be repeated for different HFTW pumping rates and regional groundwater velocities.

3.3.2 The conditions for repeated experiments

Three experiments were conducted in the artificial aquifer to ascertain the accuracy of the HFTW and transect flux measurement methods under different conditions. The conditions for each of the three experiments are shown in Table 2.

Table 2. **Experimental conditions**

Experiment	Tracer		HFTW Pumping rate (m^3/day)		Water flow rate through the aquifer (m^3/day)	Tracer injection duration
	Upflow well	Downflow well	Upflow	Downflow		
One	Bromide	Nitrate*	2.22	2.39	3.02	114 hours
Two	Nitrate**	Tritium	2.32	2.59	2.94	336 hours
Three***			2.15	2.55	3.02	

* Fluoride was also injected into the downflow well, but fluoride data were not used as it appears fluoride did not behave conservatively

** Nitrate data from experiment two were unavailable for this study

*** Only for the multi-dipole approach (No tracer injection)

3.4 Mass flux measure using the HFTW method

3.4.1 Hydraulic gradient

As described in chapter 2, mass flux can be calculated based on the values of hydraulic gradient, concentration, and hydraulic conductivity (See Eqn (2)). Hydraulic gradient *(i)* is simply the slope of the water table or potentiometric surface. It is the change in hydraulic head *(dh)* over the change in distance between two monitoring wells *(dL)*. Hydraulic head is a measure of the mechanical energy that causes groundwater to flow.

$$i = dh / dL \qquad (7)$$

Assuming homogeneity, the hydraulic heads measured at the two HFTWs will be the same value, since both wells are equidistant from the constant head boundaries at the

upper and lower ends of the artificial aquifer. In general, though, measuring the head at three wells (the two HFTWs and the observation well) will allow calculation of the magnitude and direction of the hydraulic gradient.

3.4.2 Multi-dipole approach to measure hydraulic conductivity

Goltz *et al.* (2004) presented an analytical equation to calculate drawdown resulting from operation of a multi-dipole system of wells in a horizontally infinite aquifer. The authors also developed a formula to calculate drawdown resulting from multi-dipole operation appropriate for the boundary conditions in the finite artificial aquifer (Goltz *et al.*, 2004). Using this analytical formula, if the hydrological parameters describing the system are known (well pumping rates, the hydraulic gradient, the radius and coordinates of the well, vertical coordinates of the top and bottom screens, and the thickness of the aquifer) the drawdown and mounding of the wells can be measured to allow calculation of hydraulic conductivities using inverse methods. By operating the HFTWs at a series of different flow rates, the drawdown at the downflow well and the mounding at the upflow well can be measured at each flow rate. Then the inverse methods discussed above can be applied to obtain the ¡°best¡± value of hydrauli conductivity that maximizes the objective function:

$$F_{obj} = 1 - \frac{1}{N}\sum_{i=1}^{N}\left|H_{meas}^{i} - H_{calc}^{i}\right| \qquad (8)$$

where H_{meas}^{i} and H_{calc}^{i} indicate the measured and calculated hydraulic heads at the i^{th} flow rate, respectively, and N is the total number of head measurements. The method can be applied assuming isotropic (that is, horizontal and vertical hydraulic conductivities are the same) or anisotropic conductivities.

A genetic algorithm (Carroll, 1996) will be used to determine the best value of hydraulic conductivity that maximizes the objective function.

3.4.2 Tracer test approach to measure hydraulic conductivity

When operating HFTWs, groundwater will flow from the injection screens to the extraction screens of the wells. We define interflow *(I_{ij})* as the fraction of water being drawn into extraction screen *j* that originated in injection screen *i* (Goltz *et al.*, 2004) (Figure 12). For example, I_{12} represents the fraction of water entering the lower (extraction) screen of the upflow well that originated in the upper (injection) screen of the same well. As described in Section 3.3, bromide and nitrate will be continuously added as tracer chemicals at the injection screens of the upflow and downflow wells, respectively.

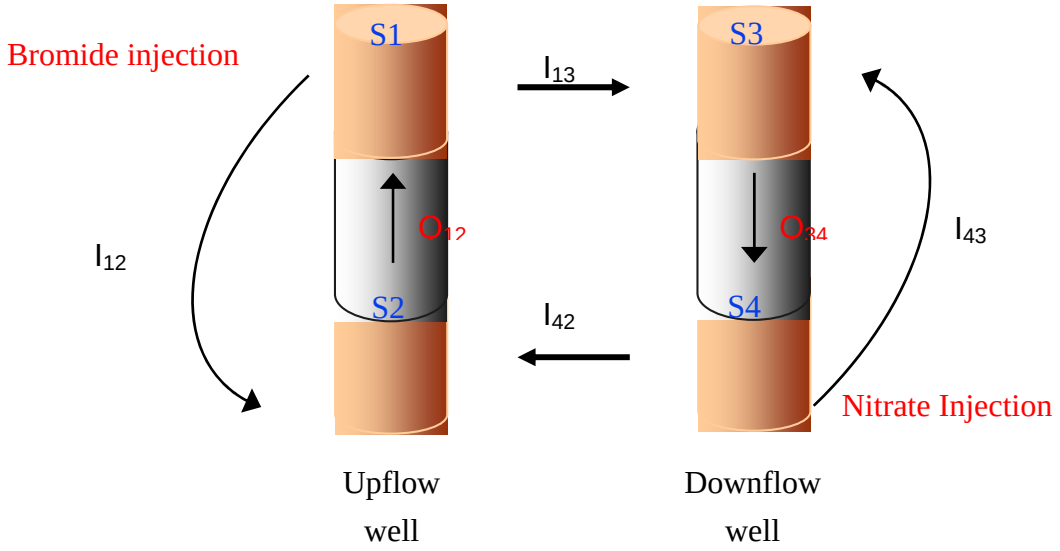


Figure 12. **HFTW interflows and tracer injection screens (Goltz *et al.*, 2004)**

Thus, if we measure the steady-state concentration of tracers in each of the four well screens, we can obtain the four interflows using the following four equations (assuming steady state and using mass balance):

$$
\begin{aligned}
B_1 I_{12} + B_4 I_{42} &= B_2 \\
N_1 I_{12} + N_4 I_{42} &= N_2 \\
B_1 I_{13} + B_4 I_{43} &= B_3 \\
N_1 I_{13} + N_4 I_{43} &= N_3
\end{aligned}
\qquad (9)
$$

where, B_i and N_i are the concentrations of bromide and nitrate measured at screen i respectively.

With an estimate of interflows based on conduct of a tracer test, inverse numerical modeling can be used to obtain hydraulic conductivity (Goltz *et al.,* 2004). Assuming a value of hydraulic conductivity, the three-dimensional numerical flow model MODFLOW (Harbaugh and McDonald, 1996) can be used to simulate interflows between the four HFTW well screens. Having values for experimentally measured and numerically calculated interflows, we can define an objective function (F_{obj}) as:

$$
F_{obj} = 1 - \frac{1}{N} \sum_{i}^{N_{inj}} \sum_{j}^{N_{ext}} \left| I_{ij}^{meas} - I_{ij}^{calc} \right| \qquad (10)
$$

where I_{ij}^{meas} and I_{ij}^{calc} are the measured and calculated interflows between injection well screen i and extraction well screen j, respectively, N_{inj} and N_{ext} are the number of injection and extraction well screens, respectively, and N is the total number of well screens.

The ¡°best hydraulic conductivity is determined when the above objective function is maximized. As with the multi-dipole technique, a genetic algorithm (Carroll, 1996) will be used to determine the best value of hydraulic conductivity that maximizes the objective function. The technique can be applied assuming both isotropic and anisotropic hydraulic conductivities.

3.4.4 Mass flux

The actual mass flux in the artificial aquifer can be known using next equation

because we know the concentration of chloride in the influent water (C), the flow rate of influent water (Q), and the cross-section area (12.22 m^2) of aquifer:

$$M_f = Q \times C \Big/ Area \qquad (11)$$

Knowing the hydraulic gradient (i) in the artificial aquifer (from Section 3.4.1), and having determined the hydraulic conductivity (K) using the methods described in Sections 3.4.2 and 3.4.3, it is only necessary to measure the chloride concentration (C) in the HFTW to obtain a measurement of chloride mass flux (M_f) using equation (12):

$$M_f = K \times i \times C \qquad (12)$$

We can now validate the HFTW method by comparing the measured mass flux to the actual mass flux. We can further compare the accuracy of the two HFTW approaches (multi-dipole vs. tracer) as well as seeing the effect of assuming hydraulic conductivity isotropy or anisotropy. Finally, we can compare the mass flux measured by the HFTW methods with the flux measured using the conventional transect method, as described below.

3.5 Mass flux measure using transect method

As explained in chapter 2.4.1, mass flux can be measured with the transect method by applying equation (2) ~ (5). For this study, we will assume the hydraulic gradient and the contaminant concentration at each sampling point are the same values as were measured in the previous HFTWs experiment.

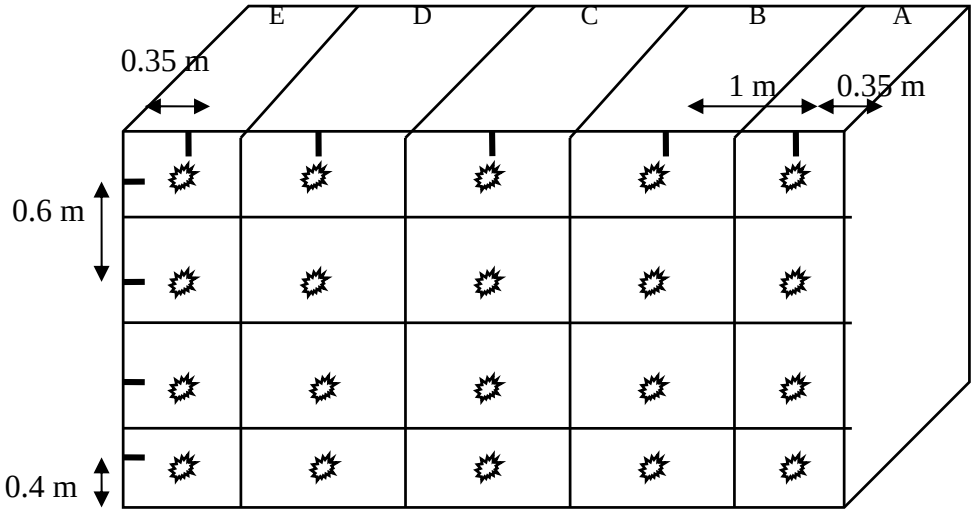


Figure 13. **Cross section view of the transect sampling points**

Hydraulic conductivities measured by Bright *et al.* (2002) at a number of the sampling points can be used in this study for measuring the mass flux using the transect method. However, because Bright *et al.* (2002) did not determine conductivities at all sampling locations, we use the quadratic Shepard method (Renka, 1998) to estimate conductivity at locations where it wasn¡¯t measure. Table 3 indicates the hydraulic conductivities that will be used in equation (2) ~ (5) to estimate mass flux. Flux will be estimated using the Table 3 conductivities at each of five transects perpendicular to the flow direction in the artificial aquifer. Bright et al. (2002) averaged these hydraulic conductivities to be a 164 *m/day* and it is well compared to the overall hydraulic conductivities calculated using flow rates, hydraulic gradients, and cross-sectional area to be 173 *m/day* and 163 *m/day*, respectively, in the experiments one and two.

Table 3. **Hydraulic conductivity of each point (*m/day*) determined by Bright *et al.* (2002). The values in the shaded boxes were estimated using the quadratic Shepard method.**

Transect (number of measured conductivities)	Row	Depth			
		0.4 m	1.0 m	1.6 m	2.2 m
1 st (19)	A	151.36	109.65	102.33	204.17
	B	151.36	131.83	83.18	93.33
	C	109.65	109.65	79.43	109.65
	D	131.83	100.00	93.05	91.20
	E	131.83	102.33	131.83	102.33
3 rd (18)	A	213.80	199.53	173.78	223.87
	B	158.49	151.36	157.63	190.55
	C	165.96	165.96	165.96	177.83
	D	165.96	165.96	169.96	190.55
	E	245.47	204.17	151.36	151.36
5 th (15)	A	288.40	147.91	138.04	138.04
	B	229.09	173.78	171.44	198.89
	C	151.36	158.49	173.78	215.69
	D	229.09	183.80	217.80	239.88
	E	263.03	151.36	173.78	154.88
7 th (11)	A	213.80	236.69	213.80	123.03
	B	194.98	193.79	172.59	131.15
	C	144.54	158.49	173.79	228.70
	D	173.78	156.33	179.72	239.88
	E	213.80	152.45	134.90	154.88
9 th (8)	A	346.74	316.20	267.17	165.96
	B	186.21	206.94	182.48	113.83
	C	478.63	276.15	257.06	263.17
	D	251.19	216.29	223.54	239.88
	E	288.40	191.41	170.95	190.55

Table 4 lists conductivities that were determined by averaging the Table 3 conductivities horizontally. Horizontal averaging is justified based on the observation that the hydraulic conductivities along the sides of the aquifer parallel to the flow direction were found to be higher than conductivities along the centerline (Bright *et al.*, 2002). The Table 4 averaged conductivities will be used to determine an average flux for the entire artificial aquifer, for comparison with the values of flux calculated at each transect.

Table 4. **Horizontally averaged hydraulic conductivity (*m/day*)**

Row	Depth			
	0.4 m	1.0 m	1.6 m	2.2 m
A	242.82	201.99	179.02	171.01
B	184.03	171.54	153.46	145.55
C	210.03	173.75	170.00	199.01
D	190.37	164.48	176.81	200.28
E	228.50	160.34	152.56	150.80

Table 5 shows the area of aquifer perpendicular to the flow direction for each sampling point for use in applying equation (3). These areas were determined by constructing Theissen polygons in the control planes.

Table 5. **Area associated with each sampling point for use in flux calculation (m^2)**

	0.4 *m*	1.0 *m*	1.6 *m*	2.2 *m*
A	0.595	0.510	0.510	0.595
B	0.700	0.600	0.600	0.700
C	0.700	0.600	0.600	0.700
D	0.700	0.600	0.600	0.700
E	0.595	0.510	0.510	0.595

3.6 Cost analysis

Cost is obviously an important consideration in deciding which flux measurement technique to apply at a site. In this section, we describe the approach that we will use to compare the costs of the four flux measurement methods (transect, PFM, IGIM, and HFTW).

To compare the methods, we will assume they are all being applied to measure the mass flux at a template contaminated site. We will define the template site as follows:

> A shallow confined sand aquifer (porosity = 0.3) contaminated with a 200 *m* wide and 10 *m* thick plume of chlorinated hydrocarbons.

The following assumptions were made:

1. Costs for manpower to operate the pumps when applying the IGIM and HFTW methods are negligible.
2. The costs for applying the two passive methods (transect and PFM) are approximately equal except for the additional cost of measuring hydraulic conductivity in the transect method. The IGIM method and the HFTW method using the tracer test

approach have increased analytical costs due to the need to conduct long-term concentration breakthrough and tracer tests, respectively.

Based on these assumptions, we list the main cost items associated with each approach in Table 6. How the unit quantities in the item description column were determined is described below.

Table 6. **Main cost items for each mass flux measurement method and approach**

Method / approach		Item Description
Transect methods		2-Inch Monitoring Wells (9 EA) Number of Contaminant Concentration Analyses (18) Pump test for measuring hydraulic conductivity (1)
PFM		2-Inch Monitoring Wells (9 EA) Number of Contaminant Concentration Analyses (18)
IGIM		8-Inch Pumping Wells (2 EA) Treatment for Extracted Contaminated Water (50,000 m^3) Contaminant Concentration Breakthrough Test Duration (9.5 days) and Number of Analyses (114)
HFTW	Multi-dipole approach	8-Inch Pumping Wells with Packers (2 EA) 2-Inch Monitoring Well (1 EA)
	Tracer test approach	8-Inch Pumping Wells with Packers (2 EA) 2-Inch Monitoring Well (1 EA) Tracer Test Duration (12.5 days) and Number of Tracer Analyses (100 per each tracer)

To estimate the number of monitoring wells to install in the transect and PFM methods, we follow Borden *et al.* (1997) and Bockelmann *et al.* (2003), who installed transect monitoring wells approximately 15 *m* ~ 40 *m* apart. Thus, for a 200 *m* wide plume, we assume 9 monitoring wells will be adequate. The number of sampling points

is determined assuming each monitoring well is constructed to sample at two depths. Also, we assume the pumping test for measuring hydraulic conductivity can be done using one of the monitoring wells.

To determine the number of IGIM wells that need to be installed, we assume each well pumps at 2500 m^3/day. A pumping rate of 2500 m^3/day was chosen based on previous applications of the IGIM at a number of sites (Table 7). The number of IGIM wells (N) and the total volume of water that needs to be extracted and treated (V_{tot}) are inversely related, as will be shown below in equation (14) below.

If r is the well capture zone radius at time t, and we need to capture a plume of width W, we find:

$$t = \frac{\pi \cdot W^2 h \cdot n_e}{4N^2 Q_{well}} \qquad (13)$$

where h, n_e, and Q_{well} symbolize the aquifer thickness, the effective porosity, and the pumping rate of each well, respectively. Thus, for our template site assumptions, with Q_{well} = 2500 m^3/day, we see that $t = 38/N^2$ *days* and we approximate that the total number of contaminant concentration measurements at a well will be $228/N^2$, if we assume an average of six measurements per day. The total volume of water that needs to be extracted and treated (V_{tot}) is:

$$V_{tot} = NQ_{well}t = \frac{\pi \cdot W^2 h \cdot n_e}{4N} \approx \frac{100{,}000 m^3}{N} \qquad (14)$$

Thus, we see that there is a cost tradeoff between the duration of the pumping test, the number of wells installed, and the volume of water that must be extracted, analyzed, and treated. If we know the unit costs for installing a treatment well (C_{well}), measuring

contaminant concentration ($C_{analysis}$), and treating the contaminated water ($C_{treatment}$), and we define n as the total number of concentration measurements made at all wells, we can determine the optimal number of wells that will result in the lowest total cost by minimizing the following objective function:

$$Total\ Cost = C_{well} \times N + C_{treatment} \times V_{tot} + C_{analysis} \times n \quad (15)$$

Subject to: $n = 6 \times t \times N = {}^{228}/_{N}$

$V_{tot} = {}^{100,000}/_{N} \quad m^3$

Using the unit costs listed in table 8, we determine that total cost is minimized for N = 2, n = 114, t = 9.5 *days*, and V_{tot} = 50,000 m^3.

Table 7. **Pumping rate, capture zone radius, and duration of pumping for IGIM application at various field sites**

Location	Pumping Rate (m^3/day)	Radius of Capture Zone (m)	Duration of Pumping (*day*)
Quaternary River Valley in Southwest Germany (Bockelmann *et al.*, 2003)	112 ~ 415	15 ~ 20	Not reported
Stuttgart (INCORE, 2003)	458	15 ~ 60	5.3
Strasbourg (INCORE, 2003)	2013 ~ 4750	18 ~ 55	3
Linz (INCORE, 2003)	1296 (Maximum)	23 ~ 46	5
Milan (INCORE, 2003)	2592 (Maximum)	29 ~ 39	7.3

We can increase the number of pumping wells to decrease the duration of pumping time, though based on the pumping durations listed in Table 7 for a number of

sites, which range from 3 ~ 7 *days*, we see that a 9.5-*day* IGIM is reasonable.

To determine the duration of the tracer test in the HFTW method, we use equation (16), to determine the minimum travel time for a tracer flowing between an injection and extraction well (t_{min})

$$t_{\min} \cong \frac{4}{3}\pi \frac{a^2 H n_e}{Q} \qquad (16)$$

where a, H, n_e, and Q symbolize the half-distance between the injection/extraction wells, the thickness of the screened section of wells, the aquifer porosity, and the wells¡¯ pumping rates, respectively (Cunningham *et al.*, 2004). Cunningham *et al.* (2004) graphed both measured and theoretical breakthrough curves. Looking at these graphs, we approximate that it requires 20 times t_{min} before tracer concentrations at the extraction screen approach steady-state. Assuming a = 5 *m*, H = 4 *m*, n_e = 0.3, and Q = 200 m^3*/day,* which is based on field data from an HFTW application (McCarty *et al.*, 1998), we find the duration of the tracer test is 12.5 *days*. Assuming an average of two tracer analyses daily for each tracer from all screens, we approximate that a total of 200 analyses (50 for each tracer) will be required.

Unit costs will be used to calculate the relative total cost of each mass flux measurement method. It is again noted that these total costs are not absolute, but relative, as the costs of items that are common to all methods are neglected. Costs for treatment and monitoring wells are based on costs at Site 19, Edwards AFB, California (AFCEE, 1998) updated to the present year assuming 3% annual inflation. It is assumed that granular activated carbon (GAC) will be used to treat the contaminated water that is extracted. Although treatment costs will vary with flow rate and concentration of contaminant, we roughly assume $1 per 1 m^3 based on Federal Remediation Technologies

Roundtable (FRTR, undated internet) data showing \$0.32~\$1.7 per m^3 at flow rates of 400 m^3/day. Costs for tracer analyses are based on costs from the Hoosier Microbiological Laboratory (HML, 2001). These costs are \$150, \$60, and \$18 for chlorinated hydrocarbon, bromide and nitrate-N analysis, respectively. The cost for a pumping test for measuring hydraulic conductivity was approximate at \$2000.

Table 8. **Unit costs for representative items**

Item		Cost	
		1998	2005
8-Inch Treatment Well (EA)		\$22,723	\$27,946
8-Inch Treatment Well with Packer (EA)		\$27,392	\$33,689
2-Inch Monitoring Well (EA)		\$13,723	\$16,878
Contaminated Water Treatment (per m^3)			\$1
Contaminant Concentration Analysis (Chlorinated hydrocarbon, per analysis)			\$150
Tracer Analysis	Bromide (per analysis)		\$60
	Nitrate (per analysis)		\$18
Pump Test for measuring hydraulic conductivity			\$2000

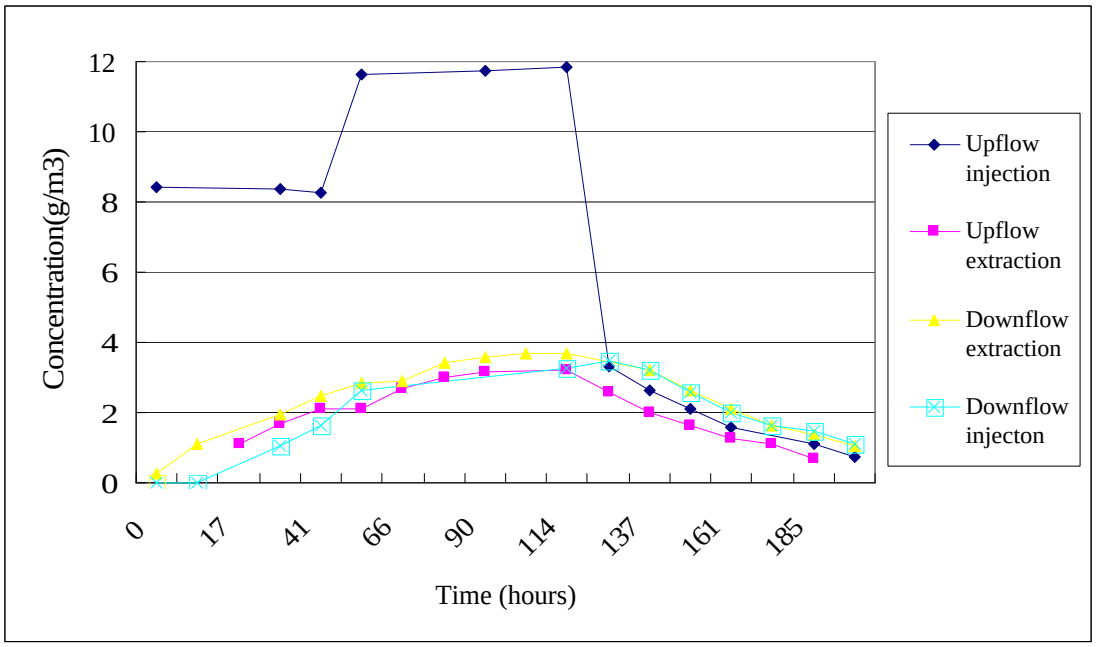
IV. Results and Discussion

4.1 Introduction

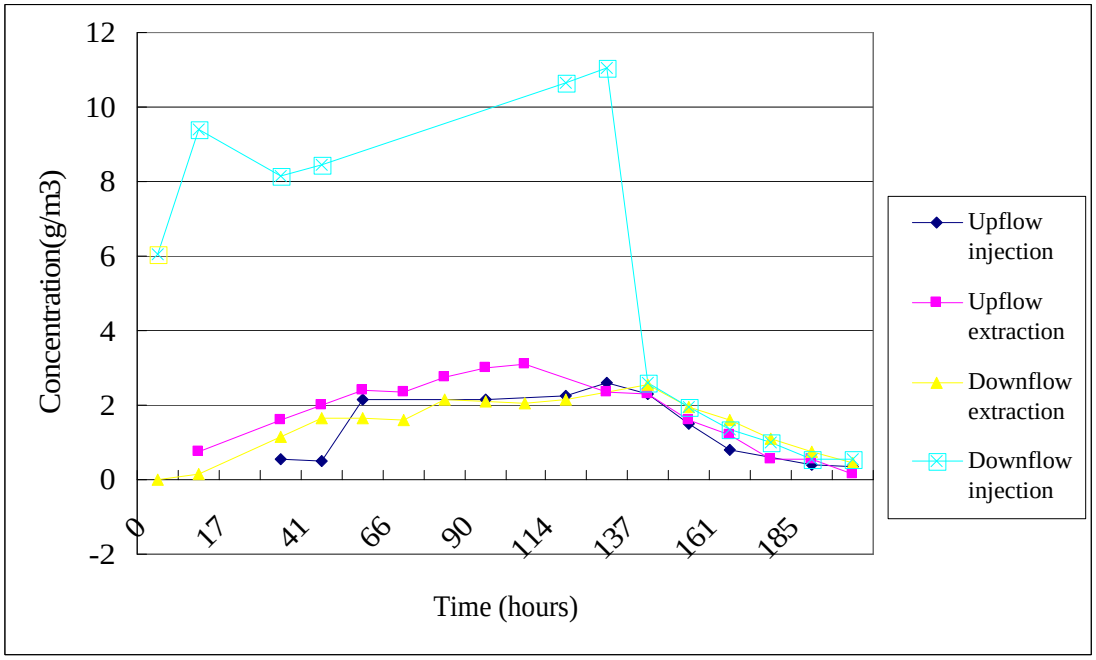
In section 4.2, data obtained from three experiments conducted in the artificial aquifer are presented. The mass fluxes measured using the HFTW method and the transect method are analyzed in section 4.3 and costs for the different flux measurement methods are calculated in section 4.4. In section 4.5 the flux measurement results are compared and discussed in light of the analyses in the previous sections.

4.2 Experimental Data

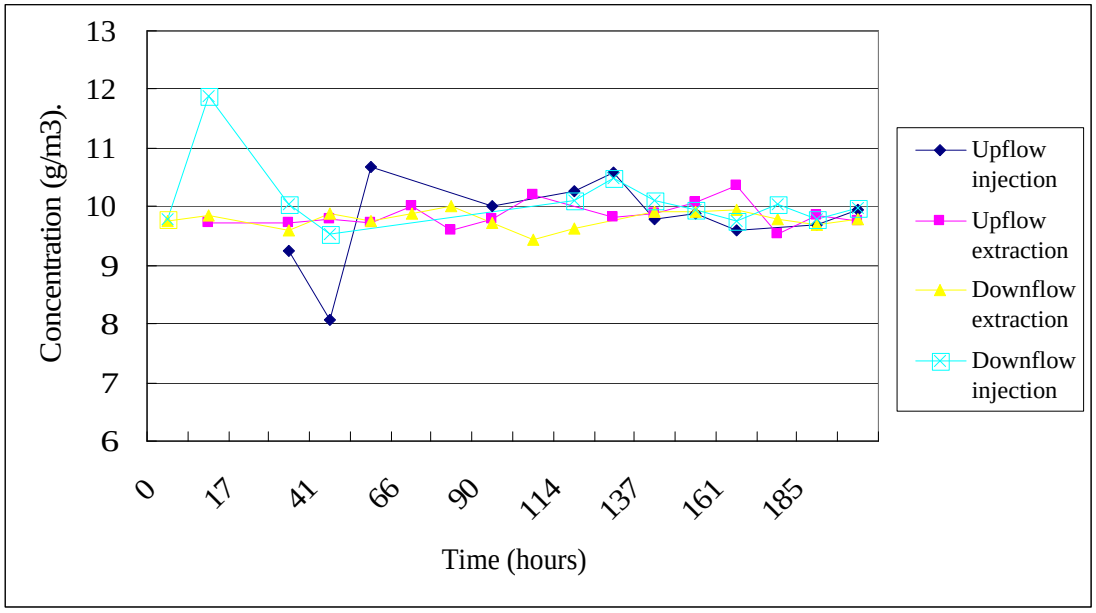
Figure 14 shows the concentration breakthroughs for chloride (Figure 14 (c)) and the two tracers (Figures 14 (a) and (b) for bromide and nitrate, respectively) at the four HFTW well screens for the first experiment (Table 2). Recall that to apply equation (9) we need to know the steady-state tracer concentrations at the well screens. Unfortunately, from Figure 14 (a) and (b), it is not apparent that steady-state has been attained in the 114 hours of tracer injection. This motivated the use of longer tracer injection duration in the second experiment. Figure 14 (c) confirms that the chloride contaminant concentration is relatively constant in time and space at 10 g/m^3.


12
10
8
6
4
2
0
Concentration(g/m3)
0
17
41
66
90
114
137
161
185
Time (hours)
Upflow injection
Upflow extraction
Downflow extraction
Downflow injecton

(a)


12
10
8
6
4
2
0
-2
Concentration(g/m3)
0
17
41
66
90
114
137
161
185
Time (hours)
Upflow injection
Upflow extraction
Downflow extraction
Downflow injection

(b)



(c)

Figure 14. **Experiment 1 concentration vs time responses at the HFTW screens: (a) bromide, (b) nitrate, and (c) chloride**

Figure 15 shows the tritium tracer concentration vs. time at each HFTW screen for the second experiment. The second experiment was conducted over a longer time frame than the first experiment in order to better establish the steady-state tracer concentration (Table 2). From Figure 15, it appears that after approximately 100 hours steady-state concentrations of the tritium tracer have been attained at the four HFTW screens. This gives us confidence that we may be able to use the later time breakthrough data from the first experiment (Figure 14) to estimate steady-state tracer concentrations at the HFTW screens.

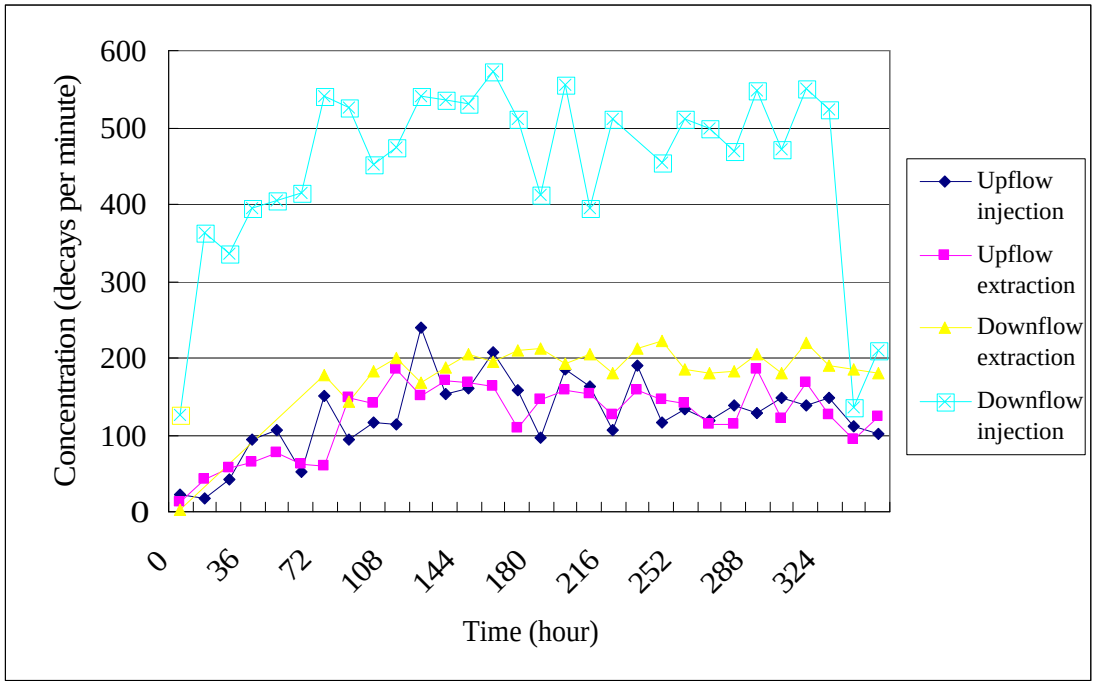


Figure 15. **Experiment 2 tritium concentration vs time responses at the HFTW screens**

During the two experiments, water head data were obtained at the HFTWs in order to apply the multi-dipole approach. In addition, a short-term third experiment was run, without tracer injection, in order to obtain additional head data that could be used for a third application of the multi-dipole approach (Table 9).

Table 9. HFTW **water head changes for application of the multi-dipole approach**

Experiment	HFTW Pumping rate (m^3/day)		Water flow rate through the aquifer (m^3/day)	Drawdown (mm)	
	Upflow well	Downflow well		Upflow well (mounding)	Downflow well (drawdown)
One	2.22	2.39	3.02	3.4	6.6
Two	2.32	2.59	2.94	8.0	7.8
Three	2.15	2.55	3.02	5.0	5.8

4.3 Mass fluxes

4.3.1 Actual mass flux

The actual chloride mass flux in the artificial aquifer for each experiment can be determined for each of the different aquifer water flow rates shown in Table 9. Applying equation (11), using a chloride contaminant concentration of 10 g/m^3 and a cross-sectional area for the artificial aquifer of 12.2 m^2, we obtain actual mass fluxes for experiments one, two, and three of 2.48 $g/m^2 \times day$, 2.40 $g/m^2 \times day$, and 2.48 $g/m^2 \times day$, respectively.

4.3.2 Application of the multi-dipole approach

Goltz *et al.* (2004) showed that using the multi-dipole approach to measure hydraulic conductivities in the artificial aquifer assumed anisotropic condition resulted in significant experimental errors. This appeared due to the small magnitude of drawdown and mounding that needed to be measured. However, Goltz *et al.* (2004) did not calculate conductivity using the multi-dipole approach assuming isotropic conditions,

which is probably a more realistic assumption for the artificial aquifer. In the current study, we use HFTW flow rates somewhat larger than the rates used by Goltz *et al.* (2004) in order to increase drawdown and mounding, and we assume isotropy, in order to apply the multi-dipole approach to estimate conductivity and flux.

Table 10 shows the best fit values of hydraulic conductivity (assuming isotropy and anisotropy) and chloride mass flux measured using the multi-dipole approach. Mass flux was calculated from the conductivity using equation 12 with a chloride concentration of 10 g/m^3 and a hydraulic gradient of 0.00143, 0.00148, and 0.00143 for the first, second, and third experiments, respectively. Inconsistently, the second hydraulic gradient is larger than the first and third hydraulic gradients even though the aquifer flow rate of the second experiment is smaller than the flow rates of the other experiments (see Table 2). This inconsistency appears to be due to experimental error in measuring the water heads. The hydraulic gradients used in this study were measured at upgradient and downgradient sampling lines, which were separated by 9.099 m. The head measurements that were used in the experiments are shown in Table 11. As the table shows, only one head measurement was recorded for Experiments 1 and 3, while there¡¯s a temporal variation in the hydraulic gradient in Experiment 2 from (0.00143 to 0.00165), which would explain the inconsistency in the aquifer flow versus hydraulic gradient measurements for the three experiments.

Table 10. **Hydraulic conductivities and mass flux measured using the multi-dipole approach**

Experiment	Hydraulic Conductivity [*m/d*]		Mass Fluxes [g/m^2*d]		
			Measured		Actual
	Anisotropic ($k_r \neq k_z$)	Isotropic ($k_r = k_z$)	Anisotropic (using k_r)	Isotropic	
One	k_r=1.57 k_z=0.1	1.13	0.022	0.016	2.48
Two	k_r=3.76 k_z=16.18	20.16	0.056	0.298	2.40
Three	k_r=16.29 k_z=22.01	16.35	0.233	0.234	2.48
Total	k_r=28.14 k_z=7.14	16.5	0.407	0.239	approximately 2.45

Table 11. **Water heads and hydraulic gradients in the artificial aquifer experiments**

Time (hr)	Water head (*cm*)				Hydraulic gradient (Experiment 2)
	Upgradient		Downgradient		
	Experiments 1 and 3	Experiment 2	Experiments 1 and 3	Experiment 2	
0	18.3	18.3	17	16.9	0.00154
22		18.3		16.9	0.00154
71.75		18.3		16.9	0.00154
97.5		18.3		17	0.00143
123		18.3		17	0.00143
145.5		18.3		17	0.00143
168		18.4		16.9	0.00165
192		18.3		17	0.00143
241		18.3		16.9	0.00154
269.5		18.4		17.1	0.00143
289		18.3		17	0.00143
312		18.4		17.1	0.00143
336		18.3		17	0.00143
408		18.4		17	0.00154
average	18.3	18.32857	17	16.97857	0.00148

For the total results in Table 10, the objective function in equation 8 was minimized by selecting values of conductivity that resulted in a best fit of model-simulated drawdown/mounding to the drawdown and mounding measurements for all three pump tests, simultaneously. The average hydraulic gradient of 0.00145 for the three experiments was used in the model. The mass flux measurements for the anisotropic condition assumption in Table 10 were calculated using the horizontal hydraulic conductivity because we can assume all water flow in the artificial aquifer is horizontal. In other applications, where this assumption may not hold, both horizontal

and vertical hydraulic conductivities may need to be considered to calculate mass flux.

We note a number of problems with the results of the multi-dipole approach presented in Table 10. First and foremost, the measured mass fluxes are one to two orders of magnitude less than the actual flux. We also see that when we assume anisotropy, vertical conductivity is determined to be larger than horizontal conductivity in experiments 2 and 3, an unlikely situation. We also note large variations in the conductivity and mass flux measurements in the three experiments. It appears that the multi-dipole approach is problematic, perhaps due to the sensitivity of the results to the relatively small drawdown and mounding that needs to be quantified. The potential for measurement error confounding results is especially apparent when we look at the head measurements in Table 9 for experiment 1. For the relatively homogeneous, confined, artificial aquifer, we would expect drawdown and mounding measurements to be approximately equal (as, indeed, they are for Experiments 2 and 3). However, in Experiment 1, we see the drawdown measurement is nearly twice the mounding measurement, indicating that measurement errors may play an important role in affecting the results of the multi-dipole approach.

4.3.3 Application of the tracer test approach

As discussed earlier, the key to applying the tracer test approach is to estimate the steady-state concentration of tracer at each of the four HFTW screens. This can be problematic, particularly for experiment 1, where steady-state concentrations were not obviously reached. In this study, we will estimate the steady-state tracer concentrations in experiment 1 using four methods, to ascertain how sensitive the hydraulic conductivity and mass flux measurements are to the method used to estimate steady-state tracer

concentration. The four estimation methods are described in Table 12.

Table 12. **Methods used to estimate steady-state tracer concentration at HFTW well screens in experiment 1 (see Figure 14)**

Estimat-ion Method	Location of Screens	Data Used for Estimate (Hrs)	Remarks
1*	- Bromide (upflow injection, downflow extraction and injection) - Nitrate (upflow injection and extraction, downflow injection)	54~125	Relatively constant over this time peirod
	Bromide (upflow extraction) Nitrate (downflow extraction)	78~125	Concentration increase at about 78 hrs
2**	Bromide (all screens) Nitrate (all screens)	114	
3*	Bromide (all screens) Nitrate (all screens)	114	
4**	Bromide (all screens) Nitrate (all screens)	Variable	Peak concentration of each tracer

* Because bromide concentrations in the extraction and injection screens of the downflow well and the nitrate concentrations in the extraction and injection screens of the upflow well should be the same, we averaged the two concentrations.
** Bromide concentrations at the downflow well screens and nitrate concentrations at the upflow well screens were not averaged as above? actual concentrations were used.

Experiment 2 appears to have attained steady-state after 108 *hours* (see Figure 15), so the tritium concentration data from 108 to 300 *hours* will be used to estimate the steady-state tritium concentration at the four HFTW screens.

Table 13. **Steady-state tracer concentrations at the HFTW screens estimated using the different approaches in Table 12.**

Experi-ment	Steady-state tracer concentration estimation method	Tracer concentration (g/m^3)							
		Bromide				Nitrate			
		Upflow		Downflow**		Upflow**		Downflow	
		injection	extraction	extraction	injection	injection	extraction	extraction	injection
One	1	11.74	3.01	3.23 (3.34)	3.23 (3.12)	2.48 (2.29)	2.48 (2.67)	2.20	10.84
	2	11.86	3.14	3.66	3.24	2.24	3.12	2.17	10.63
	3	11.86	3.14	3.45 (3.66)	3.4 (3.24)	2.68 (2.24)	2.68 (3.12)	2.17	10.63
	4	11.86	3.21	3.66	3.47	2.61	3.12	2.56	11.05
Two		Tritium concentration (decays/minute)							
		499.73*	195.25*	148.78* (147.25)	148.78* (150.31)	148.78 (150.31)	148.78 (147.25)	195.25	499.73

* Estimated from data obtained from tritium injection into the downflow well, assuming injection into the upflow well would result in a mirror image response

** Because bromide and tritium concentrations in the extraction and injection screens of the downflow well and the nitrate concentrations in the extraction and injection screens of the upflow well should be the same, we averaged the two concentrations. Numbers in parentheses indicate measured concentrations before averaging

Table 13 shows the steady-state tracer concentrations at the well screens for both experiments. For experiment 1, Table 13 shows the results calculated using each of the four steady-state concentration estimation methods discussed in Table 12. In the case of experiment 2, only the concentrations of tritium, which was injected into the downflow well, were available. Data for nitrate, which was injected into the upflow well, were unavailable. To deal with this, we assumed the nitrate breakthrough responses would mirror the tritium responses. Also note that the tritium concentrations in the extraction

and injection screens of the upflow well should be the same, and in fact, are slightly different each other. For this reason, we averaged the two concentrations as we did in methods 1 and 3 of experiment 1. This approach introduces some error, as the upflow and downflow wells had different pumping rates.

Table 14 shows the hydraulic conductivities and mass fluxes calculated using the tracer test approach with a hydraulic gradient of 0.00143 and 0.00148 respectively for the two experiments.

Table 14. **Hydraulic conductivity and mass flux calculated using the tracer test approach**

Experiment	Steady-state tracer concentration estimation method	Hydraulic Conductivities [*m/d*]		Mass Fluxes [g/m^2*d]		
				Measured		Actual
		Anisotropic (kr ¡Ákz)	Isotropic (kr = kz)	Anisotropic (using kr)	Isotropic	
One	1	kr=132 kz=46	230	1.89	3.29	2.48
	2	kr=104 kz=40	243	1.49	3.47	
	3	kr=104 kz=40	230	1.49	3.29	
	4	kr=97 kz=36	234	1.39	3.35	
Two		kr=93 kz=59	143	1.38	2.12	2.40

For the first experiment assuming isotropy, the measured mass fluxes are relatively consistent, with values that overestimate the actual mass flux between +33% and +40%, with the average of the four measurements overestimating flux by 35%.

Apparently, at least for the assumption of isotropy, the mass flux measurement is not very sensitive to the method used to estimate the steady-state tracer concentration at the HFTW screens. Assuming anisotropy, the mass flux measurements were somewhat more variable, ranging between -44% and -24% from actual values, with the average of the four measurements underestimating flux by 37%. When assuming anisotropy, we can see the horizontal hydraulic conductivities are larger than the vertical hydraulic conductivities, as would be expected.

For the second experiment, we underestimate mass flux whether we assume isotropy (14% underestimate) or anisotropy (44% underestimate). Similarly, Goltz *et al.* (2004) found that application of the tracer method resulted in an underestimate of hydraulic conductivity, which would lead to an underestimate of mass flux, of 6% (assuming isotropy) and 24% (assuming anisotropy).

It appears that for the relatively homogeneous and isotropic artificial aquifer, the mass fluxes measured by the HFTW method when assuming isotropy are better (considering both accuracy and consistency of results for different averaging techniques) than those measured assuming anisotropic conditions.

We note that the flux estimated assuming anisotropy is consistently 40% ~ 65 % less than the flux estimated assuming isotropy. Similarly, Goltz *et al.* (2004) found that the hydraulic conductivity obtained assuming anisotropy was less than the conductivity obtained assuming isotropy.

4.3.4 Application of the transect method

Hydraulic conductivities for each sampling well in the artificial aquifer are listed in Table 3, horizontally averaged hydraulic conductivities are listed in Table 4, and the

areas associated with each sampling point are listed in Table 5. Based on the respective hydraulic gradients of 0.00143 and 0.00148 for experiments one and two and the chloride concentration of 10 g/m^3, we can apply equations (2) ~ (5) to calculate mass flux through each transect (Table 15). Also, if we assume it¡¯s appropriate to use horizontally averaged hydraulic conductivities (Table 4) we can calculate an overall mass flux through the artificial aquifer (Table 16).

Table 15. **Mass flux through each transect measured using the transect method (see Table 3)**

Experiment	Transect row	Number of Points at which Conductivity Measured	Number of Points at which Conductivity Estimated using Shepard Method	Mass Flux [g/m^2*day]	
				Measured	Actual
One	1	19	1	1.66	2.48
	3	18	2	2.57	
	5	15	5	2.75	
	7	11	9	2.57	
	9	8	12	3.47	
Two	1	19	1	1.72	2.40
	3	18	2	2.66	
	5	15	5	2.85	
	7	11	9	2.66	
	9	8	12	3.59	

Table 16. **Mass flux measured using transect method with horizontally averaged hydraulic conductivities (see Table 4)**

Experiment	Number of Conductivity Measurements	Mass Flux [g/m^2*day]	
		Measured	Actual
One	71	2.60	2.48
Two	71	2.70	2.40

From Table 15 we can see that there seems to be no relationship between the number of conductivity measurements made in a row and the accuracy of the flux measurement and that fluxes measured in the first and ninth rows are significantly different from both the actual flux and the flux measured in rows 3, 5 and 7. The fluxes measured in rows 3, 5, and 7 are relatively consistent, and are close to the actual flux value. Since the hydraulic gradient is assumed constant throughout the aquifer, the difference in the fluxes measured in rows 1 and 9 is a direct result of the fact that the hydraulic conductivities measured in those rows by Bright *et al.* (2002) (Table 3) are significantly different than the conductivities measured in the other rows. It would be necessary to measure hydraulic gradient at locations throughout the aquifer to obtain an estimate of flux through each of the rows using the transect method. Also note that the difference in measured fluxes in experiments one and two is strictly due to the difference in hydraulic gradients in the two experiments, since the conductivities used to calculate flux were the same for both experiments.

The mass fluxes measured for both experiments using the horizontally averaged conductivities are slightly overestimated from the actual fluxes (+5 % for experiment one and +12 % for experiment two).

4.4 Cost

Using the primary cost drivers (Table 6) and unit costs (Table 8) of the measurement methods, along with the cost estimation methods described in Chapter 3, we calculate the relative costs of applying each of the flux measurement methods at a template site (Table 17). Note that these costs are intended for comparison purposes only, and costs common to the methods have been omitted from the analysis.

Table 17. **Relative costs of applying the different mass flux measurement methods at a template site**

Method / approach		Relative Cost
Transect		$156,602
PFM		$154,602
IGIM		$122,992
HFTW	Multi-dipole approach	$84,256
	Tracer test approach	$92,056

Table 17 shows that both HFTW approaches are much cheaper than the other three methods at our template site. The number of monitoring wells required for the transect and PFM methods are a significant expensive, while the cost of water treatment to apply the IGIM controls the cost of that method. The transect method is more expensive than the PFM method, largely due to the need to conduct a pump test to measure hydraulic conductivity when applying the transect method. The cost of the IGIM method is very dependent on the scale contamination because this method measures mass discharge and therefore requires that the entire plume be captured. The costs of other three methods are less dependent on the scale of the contamination as they

can be applied to quantify flux in a representative cross-sectional area of the plume without requiring capture of the entire plume.

4.5 Overall comparison of methods

4.5.1 Accuracy

Mass fluxes measured in section 4.3 are compared to the actual mass flux and the errors between measured and actual mass fluxes tabulated in Table 18.

Table 18. **Measured mass flux error for each method**

Exper-iment	HFTW					Transect	
	Multi-dipole		Tracer Test				
	An-isotropy	Isotropy	Method	An-isotropy	Isotropy	Transect Number	Results
One	-99 %	-99 %	1	-24 %	33 %	1	-33 %
			2	-40 %	40 %	3	3 %
			3	-40 %	33 %	5	11 %
			4	-44 %	35 %	7	4 %
						9	40 %
						Averaged	5 %
Two	-98 %	-88 %	-	-44 %	-14 %	1	-28 %
						3	11 %
						5	19 %
						7	11 %
						9	50 %
						Averaged	12 %
Three	-91 %	- 91 %	-	-	-	-	-
Total	-84 %	-90 %	-	-	-	-	-

We see from Table 18 that the multi-dipole approach of the HFTW method results in significant flux underestimates. It appears the method is overly sensitive to the relatively small values of drawdown and mounding that are observed at the HFTWs, at

least for the conditions of the artificial aquifer, where well pumping rates on the order of 2-3 m^3 *per day* result in water level changes on the order of millimeters.

On the other hand, application of the HFTW method using the tracer test approach demonstrated that mass flux can be measured within about ±44%. The results showed that at least for the relatively homogeneous and isotropic artificial aquifer, mass fluxes estimated assuming anisotropy are consistently less than mass fluxes estimated assuming isotropy.

The transect method also resulted in flux estimates that were within about ±50% of the actual flux. In the case of the artificial aquifer, where flow is horizontal, it was also seen that horizontally averaging hydraulic conductivities over multiple transects resulted in a flux estimate that was quite accurate (within 15% of the actual value).

4.5.2 Other considerations

Table 19 qualitatively compares the different methods in terms of cost (based on Table 17), accuracy (based on Table 18), and other considerations which are discussed below.

Table 19. **Comparison of Flux Measurement Methods**

	Transect Method	PFM	IGIM	HFTW Tracer Approach*
Cost	Poor	Poor	Moderate	Good
Accuracy	Moderate	-	-	Moderate
Simplicity/Implementability	Good	Moderate	Moderate	Poor
Regulatory Considerations	Good	Moderate	Good	Poor
Availability	Good	Poor	Moderate	Poor

* Due to its poor accuracy, the HFTW multi-dipole approach is not considered in this comparative analysis

4.5.2.1 Simplicity / Implementability

The conventional transect method, which consists of taking hydraulic gradient measurements, installing and sampling monitoring wells, and conducting a pump test to determine hydraulic conductivity, is simple to implement. Each step in the method is well-understood and easy to apply. The PFM, IGIM, and HFTWs methods are somewhat more complex. The PFM method requires quantifying the contaminant sorbed onto the permeable sorbent, as well as measuring the loss of resident tracer. Both of these measurements, as well as their interpretation, require special expertise. The IGIM requires installation of pumping wells that will capture the contaminant plume. Thus, considerable site characterization is required, to determine the location and pumping rates of the wells in order to capture the plume. In addition, interpretation of the concentration breakthrough data at each of the pumping wells is somewhat complex (Bockelmann et al., 2001). Finally, the HFTW wells are specially constructed dual-screened wells with a packer to separate the upper and lower well screens. The downflow well requires special construction to pump in a downwards direction. Thus, implementation of the method is somewhat difficult.

4.5.2.2 Regulatory Considerations

Both the HFTW tracer test approach and the PFM method involve injecting tracers into the aquifer, and this may raise regulatory concerns. In addition, the HFTW method involves circulating contaminated groundwater in the subsurface. If contaminant concentrations vary in space (particularly vertically) this may also concern regulators. Both the IGIM method and the pump test portion of the transect approach are of some small concern since they require contaminated groundwater extraction and

treatment.

4.5.2.3 Availability

The transect approach is well-understood, has appeared many times in the literature, and involves no special expertise. Therefore, it is readily available from any purveyor of groundwater remediation services. The other methods are all in some stage of technology transfer, with the IGIM method furthest along, followed by the PFM method and then the HFTW method. The IGIM method could probably be applied by well-trained practitioners who are familiar with the appropriate literature. However, the PFM and HFTW methods are unavailable for field application? their use at a site would require the assistance of the technology developers.

V. Conclusions

5.1 Summary

In this study, we began by demonstrating the need for groundwater contaminant mass flux measurements in order to prioritize site cleanups, evaluate the efficacy of remediation technologies, estimate the rate of natural attenuation of contaminants, and develop a source term for application in contaminant transport models.

Four methods of measuring mass flux that have appeared in the literature were discussed: (1) the conventional transect method, (2) the integral groundwater investigation method (IGIM), (3) the passive flux meter (PFM) method, and (4) the horizontal flow treatment well (HFTW) method. This thesis focused on validating the HFTW method using data from an artificial aquifer, where mass flux was known. Results of HFTW flux measurements were also compared with flux measurements obtained from the conventional transect method.

Two approaches that had been proposed for applying the HFTW method were investigated; the multi-dipole and tracer-test approaches. Using the artificial aquifer, head data were obtained in three experiments for application of the multi-dipole approach. Two tracer tests were also run in the same artificial aquifer in order to apply the tracer test approach. Simultaneously, measurements of the hydraulic gradient were used in conjunction with previous hydraulic conductivity and concentration measurements in order to apply the transect method.

Finally, all the flux measurement methods were compared with respect to accuracy, cost, and other considerations relevant to their application at contaminated sites.

5.2 Conclusions

Although inexpensive, the multi-dipole approach of the HFTW method had large errors, raning from -84 % ~ -99 % of the actual value. Results obtained using the approach were extremely sensitive to head measurements in the wells. As head differences were on the order of millimeters, obtaining an accurate estimate of flux was difficult.

The tracer-test approach of the HFTW method measured flux within 44 %. Results obtained using the approach were relatively insensitive to the method used to interpret the tracer test data.

The conventional transect method measured flux within Z 50 % of the actual value. It should be noted, though, that this level of accuracy required use of the very dense network of monitoring wells found in the artificial aquifer.

For comparison purposes, the PFM method measured flux within 17 % in studies using laboratory columns and small-scale artificial box aquifers (Hatfield *et al.,* 2001; Hatfield *et al.,* 2004; Jonge and Rothenberg, 2005). There have been no reports of the IGIM accuracy, as it has thus far only been applied in the field, where the actual flux is not known. A direct comparison of the accuracy of PFM and HFTW methods is not possible, due to the different measurement scales in this study and the studies reported in the literature.

From the cost analysis we determined that in relative terms the HFTW method (both the multi-dipole approach and the tracer test approach) is the most economical mass flux measurement method, while the PFM and transect methods are the most expensive.

With regard to other qualitative factors such as simplicity and implementability,

regulatory concerns, and availability, the HFTW method, which is an innovative, untested approach, has many disadvantages while the conventional transect method has the most advantages.

Assuming many of the concerns regarding the HFTW method (e.g. availability, implementability) will be allayed as it progresses beyond the research stage, it appears the method, based upon accuracy and cost, has a great deal of potential. In particular, because the method is an integral method, it is advantageous when applied to heterogeneous sites. The current study looked at application of the HFTW method in comparison to the transect method in a homogeneous artificial aquifer. Presumably, when the methods are compared under more realistic heterogeneous conditions, the advantages of the HFTW method, with regard to both cost and accuracy, will increase.

The IGIM is best applied in an aquifer with high conductivity across a narrow, shallow contaminant plume (to minimize pumping costs). The transect and PFM methods have advantages when conditions are relatively homogeneous, and the plume is relatively shallow. The HFTW method has advantages when applied to a deep plume (as pumping to the ground surface is not required), and since it¡¯s an integral method, it may be applied under heterogeneous conditions. Ultimately, a site manager should decide on an appropriate flux measurement method depending on the conditions of the site and the accuracy required.

5.3 Recommendations

1. Although the HFTW method¡¯s multi-dipole approach proved highly inaccurate, this approach perhaps should not be abandoned, as it does not have the costs and regulatory problems associated with conducting a long-tem tracer test. Further testing of the multi-

dipole approach, with higher HFTW pumping rates, may be worthwhile. It is hoped the increased pumping rates would result in more accurate measures of drawdown and mounding, which should produce improved flux measurements.

2. The nitrate tracer data from experiment 2 were not available for this study. When those data are available, they should be analyzed to determine how they affect the experiment 2 flux measurements.

3. The experiments conducted in this study involved HFTWs oriented perpendicular to the regional groundwater flow direction, pumping at rates that were not significantly different. Further tests should be conducted where pumping rates, regional gradients, and well orientation with respect to the regional gradients vary significantly, in order to determine how robust the method is.

4. Ultimately, a field validation of all flux measurement methods in a real, heterogeneous system should be conducted. This would involve application of the methods at a field site where contaminant mass flux is known, and mass balance is obtained. That is, contaminant would be injected into the aquifer at a known rate, the flux of the contaminant as it is transported through the aquifer would be measured, and then the contaminant would be captured by downgradient extraction wells and quantified, to obtain mass balance. This would allow direct comparison and quantification of the accuracy of the different flux measurement methods.

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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 074-0188

1. REPORT DATE *(DD-MM-YYYY)*	**2. REPORT TYPE**	**3. DATES COVERED** *(From ? To)*
21-03-2005	**Master¡¯s Thesis**	Sep 2003 ? Mar 2005

4. TITLE AND SUBTITLE

Validation of an Innovative Groundwater Contaminant Flux Measurement Method

5a. CONTRACT NUMBER
5b. GRANT NUMBER
5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)

Kim, Seh J., Capt, Republic of Korea Army (ROKA)

5d. PROJECT NUMBER
5e. TASK NUMBER
5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(S)
Air Force Institute of Technology
Graduate School of Engineering and Management (AFIT/EN)
2950 Hobson Way
WPAFB OH 45433-7765

8. PERFORMING ORGANIZATION REPORT NUMBER

AFIT/GES/ENV/05M-02

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
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10. SPONSOR/MONITOR¡¯S ACRONYM(S)

11. SPONSOR/MONITOR¡¯S REPORT NUMBER(S)

12. DISTRIBUTION/AVAILABILITY STATEMENT
APPROVED FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED.

13. SUPPLEMENTARY NOTES

14. ABSTRACT

Flux measurement methods can be categorized as either point methods or integral methods. As the name suggests, point methods measure flux at a specific point or points in the subsurface. To increase confidence in the accuracy of the measurement, it is necessary to increase the number of points (and therefore, the cost) of the sampling network. Integral methods avoid this disadvantage by using pumping wells to interrogate large volumes of the subsurface. Unfortunately, integral methods are expensive because they require that large volumes of contaminated water be extracted and managed. HFTWs combine the advantages of each of the two approaches described above; that is, it¡¯s anintegral technique that samples a large volume of the subsurface while not requiring extraction of contaminated water from the subsurface.

In this study, the accuracy of the HFTW flux measurement method was quantified by applying the method in an artificial aquifer, where the flux being measured was known. Two HFTW approaches, the multi-dipole approach and the tracer test approach, were compared to each other, as well as being compared to the transect method of measuring flux, which is the conventionally used point method.

Results found that the transect and HFTW tracer test approaches provided reasonably accurate measures of flux (within ±50% and ±44% respectively) in the artificial aquifer, while the multi-dipole approach was too sensitive to small hydraulic head measurement errors to be useful. A comparison of the costs of applying the different methods at a generic site showed that the HFTW method had significant cost advantages.

15. SUBJECT TERMS
Groundwater Contamination, Mass Flux, Mass Discharge, Transect Method, Passive Flux Meter (PFM) Method, Integral Investigation Groundwater Investigation (IGIM) Method, Horizontal Flow Treatment Wells (HFTW) Method

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON**
REPORT	**ABSTRACT**	**c. THIS PAGE**			Mark. N. Goltz, ENV
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Standard Form 298 (Rev: 8-98)
Prescribed by ANSI Std. Z39-18