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The effect of transient flow on contaminant dispersion in porous media

by

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in partial fulfillment of the requirements for the degree of

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"One must, however, note that the flow does not always remain the same. Thus when there are rains the flow is increased, for the water on the hills being in excess is more violently squeezed out. But in times of dryness the flow subsides because no additional supply of water comes into the spring. In the case of the best springs, however, the amount of flow does not contract very much [...] It is also necessary to find the speed of the flow, for the swifter the flow the more water the spring delivers, and the slower it is, the less."

– Dioptra, Hero of Alexandria (ca. A.D. 65)

Abstract

Our ability to predict solute transport in groundwater is limited by our imperfect understanding of the physical processes governing the spreading of underground contaminant plumes beneath the surface. Inaccurate prediction of solute migration can in turn result in unreliable risk analyses, or higher costs for groundwater decontamination. It is generally accepted that spatial variations in the hydraulic conductivity of porous materials largely contributes to the spreading of solutes dissolved in groundwater. Unsteady hydraulic gradients can also enhance this dispersion by imposing an additional source of variability on the flow field. Most field and numerical studies assume steady state groundwater flow, despite compelling field evidence suggesting that flow transience may be ubiquitous. This study characterizes the effects of transient groundwater flow on contaminant migration in both homogeneous and heterogeneous porous media.

The macroscopic dispersion of miscible solutes subjected to unsteady flow fields is assessed quantitatively through a series of laboratory experiments and numerical simulations. An innovative laboratory model is presented, which consists of a two-dimensional flow cell and coupled hydraulic control system that allow the construction of spatially homogeneous or heterogeneous porous media of prescribed statistical properties, and to impose deterministic flow transients on the system. A monitoring procedure combining image processing with spatial moment analysis is used to characterize with great spatial and temporal resolution the evolution of contaminant plumes, as measured from sequences of digital images acquired during the course of laboratory experiments.

Results suggest that the influence of flow transience on solute dispersion compares well with results reported in the literature, based on theoretical or numerical investigations. Changes in the mean flow direction significantly increase transverse dispersion in proportion to the rotation angle; conversely, longitudinal dispersivity decreases in response to variations in the flow direction, but to a lesser extent. Reversing hydraulic gradients can cause a reduction in the plume extents, or plume “shrinking”. Although both the spatial and temporal variability enhance solute spreading, heterogeneity of the porous medium can mask the temporal variations in the flow field. The increased complexity introduced by the spatial and temporal variability can lead to inconsistencies between experimental and numerical models.

Sommaire

Les processus physiques gouvernant l'étalement des panaches de contaminants présents dans l'eau souterraine ne sont qu'imparfaitement connus, ce qui limite la capacité que nous avons à prédire le mouvement de solutés dissous. Les prédictions erronées du transport de contaminants peuvent affecter la fiabilité des analyses de niveau de risque, ou augmenter les coûts reliés à la décontamination des eaux souterraines. Les variations dans l'espace de la conductivité hydraulique sont en grande partie responsables de l'étalement des substances dissoutes dans l'eau souterraine. Par ailleurs, les variations du gradient hydraulique peuvent aussi augmenter la dispersion, en imposant au champ d'écoulement une source accrue de variabilité. La majorité des études de terrain traitent l'écoulement de l'eau souterraine comme étant à l'état stable, malgré les nombreux indices qui laissent croire que l'écoulement transitoire constitue la norme plutôt que l'exception. La présente étude s'intéresse aux effets sur le transport de contaminants sujets à l'écoulement transitoire, tant dans des milieux poreux homogènes qu'hétérogènes.

La dispersion macroscopique de substances solubles, lorsqu'elles sont soumises à un champ d'écoulement transitoire, est analysée de façon quantitative à l'aide d'une série d'expériences en laboratoire et de simulations numériques. Un modèle physique innovateur est présenté, composé d'une cellule d'écoulement bidimensionnelle couplée à un système de contrôle du gradient hydraulique. Ce modèle permet de construire des milieux poreux homogènes ou hétérogènes, et d'imposer un écoulement transitoire au système. L'évolution des panaches de contaminants en laboratoire est capturée avec grande précision, tant dans l'espace que dans le temps, à l'aide d'images numériques, et grâce à une procédure combinant le traitement de l'image à l'analyse des moments statistiques.

Les résultats concernant l'influence de l'écoulement transitoire sur la dispersion de solutés sont comparables avec les résultats publiés dans le cadre d'études théoriques et numériques. Les changements de direction dans l'écoulement augmentent la dispersion transverse de façon proportionnelle à l'angle de rotation; par contre, la dispersivité longitudinale diminue, mais à un moindre degré, lorsque la direction de l'écoulement varie. Les panaches peuvent rétrécir lorsque soumis à un régime où la direction de l'écoulement est inversée. Bien que les deux sources de variabilité – dans l'espace et dans le temps – tendent à augmenter la dispersion de solutés, l'hétérogénéité du milieu poreux peut masquer les changements dans le temps du gradient hydraulique. La complexité accrue résultant de la variabilité dans l'espace et dans le temps peut produire des résultats discordants entre les modèles expérimental et numérique.

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Dedication

À ma tendre épouse, Chantalle

La patience et la compréhension dont tu as fait preuve au cours de ces longues années n'ont d'égal que ton amour. Sans ton appui inconditionnel cette thèse n'aurait sans doute jamais vu le jour. Pour les sacrifices que tu as faits, et ils furent nombreux, et pour avoir su m'épauler, surtout dans les moments les plus creux, je te dis merci.

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CHAPTER 1 – Introduction

“The good quality of water is one of things which contribute the most to the health of citizen of a city. There is nothing that the magistrate should have more of interest in than maintaining the quality of that which is useful for the drink commune of man and of animal, and remedies accidents by which these water can be altered, be in the read of fountain, of river, of brook or they run, be in the place or be conserveed that one in derive, be finally in the well of or be born of source.”

– De Jussieu, 1733

Water sustains life.

Our concern for the protection of water resources is not a contemporary trend. This was already apparent from the above quote taken from Henry Darcy’s famous work in “Appendix – Note D: *Determination of the laws of water flow through sand.*” [Darcy, 1856]. In his capacity as Inspector General of Bridges and Roads, Darcy clearly saw that understanding the physical principles that govern groundwater flow through porous media was fundamental in order to protect the quality of drinking water for the citizens living in the City of Dijon, France. Almost 150 years later, groundwater professionals routinely use, and in some cases build upon, Darcy’s Law as the foundation to better understand the motion of groundwater and protect it as a resource, or to track the motion of the dissolved solutes it carries.

Groundwater quality has become an increasingly popular topic over the last 20 or 30 years. Our ability to predict solute transport in groundwater is limited by our imperfect understanding of the physical processes governing the spreading of underground contaminant plumes beneath the surface. Inaccurate prediction of solute migration can in

turn result in unreliable risk analysis studies, or in higher costs for groundwater decontamination.

This chapter places the issue of groundwater contamination into perspective, in particular when the spreading behaviour of contaminants is influenced by spatial and temporal variability in pore-water velocities. Chapter 2 summarizes the experimental approach used to address the objectives of this work. Experimental and numerical results conducted in homogeneous and heterogeneous porous media are then presented and discussed in Chapter 3 and Chapter 4, respectively. Finally, Chapter 5 summarizes the main conclusions that stem from these observations, discusses their practical implications and proposes future directions in the wake of this work. To improve readability, the accompanying appendices present the details of the methodology. They include (A) *Laboratory Apparatus*, (B) *Image Processing*, (C) *Spatial Moments*, (D) *Macrodispersivity Calculations*, (E) *Reynolds Number*, (F) *Wall Effects*. Finally, Appendix G presents all *Experimental Results* in the form of time plots of the moment-derived quantities and resulting macrodispersivity estimates.

1 BACKGROUND

Understanding complex physical processes such as dispersion is essential to improve our ability to predict contaminant movement in porous formations. Accurate prediction of the migration in porous media of a spreading contaminant plume is a very difficult task, mainly because hydrodynamic dispersion is a non-linear process, and varies with problem scale [Gelhar *et al.*, 1992]. Spatial heterogeneity of the hydraulic properties further complicates the prediction of solute dispersion in porous media, because of the strong effect it has on dispersive processes. Pore-scale dispersion coefficients, for example, may be orders of magnitude smaller than when they are measured in the laboratory using geologic materials of similar characteristics. This scale-dependent behaviour is also apparent when laboratory and field scale measurements of dispersion are compared [Gelhar *et al.*, 1992].

Extensive large-scale data sets are necessary in order to test the theories that predict dispersion in porous media. Few such field studies have been performed in the past,

however, because of the extensive resources required for their execution. In a comparative review of data sets from 59 field sites classifying the data in terms of their reliability, *Gelhar et al.* [1992] found only 14 to be highly reliable. Even where field-scale tracer test results are available, solute transport parameters such as dispersivity and distribution coefficients may differ significantly between two field sites in close proximity. For example, *Rivett et al.* [1994a] compared macrodispersion and retardation factors at two natural gradient tracer test sites located 150 metres apart, and showed that significant differences can exist between the transport parameters from the two locations, even though they were in close proximity. The scarcity of reliable field data sets partly explains the ever-increasing popularity of various theoretical models used to study groundwater flow and contaminant transport processes. Results obtained from these models are often essential in guiding field experiments.

Traditionally, investigators have treated groundwater flow as a steady state phenomenon, and this practice is still widespread. This assumption is often necessary because of the limited amount of physical evidence that can be collected at a given site. Extensive sampling networks and prolonged sampling surveys are necessary to fully characterize the spatial and temporal nature of hydrogeologic parameters. However, most of the few (large-scale) field investigations that have amassed sufficiently detailed datasets – in terms of both spatial and temporal resolution – have reported significant transience in groundwater flow conditions. This suggests that most sites may in fact exhibit transient groundwater flow regimes, and that steady state flow might in fact constitute a rather rare occurrence. The high price tag associated with such studies explains why few data sets are available that are comprehensive enough to establish a detailed configuration of flow over time. Recent detailed field investigations include the Canadian Forces Base (CFB) Borden site, near Alliston, Ontario [*Mackay et al.*, 1986], Cape Cod, Massachusetts [*LeBlanc et al.*, 1991], and the Columbus Air Force Base site, Mississippi [*Boggs et al.*, 1992]. Results from these field studies have shown temporal variations in the flow velocity and/or direction over time.

Laboratory (i.e. physical) models are a convenient compromise between numerical models and field studies. They allow various processes to be investigated under

controlled conditions, and provide valuable insight to assess and refine the theoretical models. Laboratory models are also an economical alternative to field investigations, although their usefulness is often limited by the unavoidable simplifying conditions necessary in their construction. Therefore, the main challenge facing the experimenter is to incorporate into the physical model as much of the “real life” complexities observed in the field as is possible given the technical constraints of the laboratory.

1.1 Groundwater contamination

The most effective methods for aquifer protection are undoubtedly those that prevent groundwater contamination. Unfortunately, many aquifers (most of which are located in densely populated areas) have already been exposed to pollutants. The extensive use of petroleum products and other organic chemicals in the past half-century, combined with an obvious lack of early legislative control and waste management programs, has led to many cases of such compounds being introduced into the environment. A large number of near-surface aquifers have been contaminated by human activity. Pollution sources include, to name a few, accidental above- and below-ground spills, improper waste disposal, and underground constructions such as pipelines and storage tanks which leak due to rupture, corrosion or defects. In industrialized countries, hundreds of thousands of steel gasoline storage tanks are buried in the ground. In the United States alone, an estimated 300,000 to 400,000 sites are thought to contain contaminated groundwater [National Research Council, 1994]. Petroleum products are distributed routinely via underground pipelines over many thousands of kilometres or are transported by land and sea everywhere around the globe. Leakage and accidental spills from all these sources represent an ever-increasing threat to groundwater quality [Freeze and Cherry, 1979].

A large proportion of groundwater contaminants are organic chemicals. These include, for example, gasoline (composed of hundreds of individual compounds), polychlorinated biphenyls (PCBs), and dry-cleaning solvents. Mackay and Cherry [1989] compiled data from selected contaminated sites organic contaminant plumes. The volume of contaminated water at these sites was estimated to represent between 400 000 and 56 million times the actual volume of contaminant that had originally been introduced into

the subsurface. This can be explained by the dispersive nature of the motion of fluids through porous media and by the fact that organic compounds typically exhibit extremely low solubility in water. But despite the diluted nature of these plumes, they pose a significant health and environmental risk because they are also considered toxic at very low concentrations.

1.2 Groundwater remediation

A great deal of money and effort have been – and continue to be – devoted to aquifer restoration in an attempt to return contaminated sites to their original unpolluted state, or at a minimum to meet the decontamination objectives set forth by the legislation. The much-diluted nature of organic contaminants explains why the more conventional approaches, such as containment or contaminant withdrawal, are in general relatively successful at removing the product found as a separate (immiscible) phase, but only a fraction of the adsorbed and dissolved phases. For the non-aqueous phase liquids (NAPL) that remain trapped in the soil matrix where they act as a quasi-permanent source of residual contamination, or for the fraction of free product that cannot be recovered (in isolated pools, for example), the mass of contaminant must generally be removed in a much more dilute form. This can be achieved, for example, by vaporization into the soil gas or by dissolution in groundwater [*Mackay and Cherry, 1989*]. These mass transfer processes generally take place at a slow rate, however, and various ways of enhancing them have been proposed and tested in recent years, most showing marginal success at best.

The basis for remediation at most contaminated sites is to extract the water, treat it by stripping the contaminants from the water, and re-inject the treated water or dispose of it in some other way [*Mackay and Cherry, 1989*]. This general approach is commonly referred to as pump-and-treat (PAT) remediation. Removal of dissolved contaminants by pumping requires the extraction of a quantity of water much larger than the actual volume that is contaminated [*Mackay and Cherry, 1989*]. For example, *Rivett et al. [1992]* estimated that a volume 15 times the size of the original TCE-contaminated zone had been pumped at a field study site after only 100 days of pumping. Pumping operations

had to be continued for a further 300 days before the contaminant concentrations dropped near drinking water standards. This is partly due to the fact that the contaminants are retarded due to sorption, and move more slowly than groundwater. In addition, the contaminant may be poorly soluble in water (as is the case for most organic compounds). If such is the case, it is generally observed that it will remain trapped in the porous matrix, where it will dissolve slowly into groundwater, acting as a long-term source of pollution. As a result, the PAT approach may utilize a considerable volume of uncontaminated groundwater surrounding the site to flush the contaminants from the polluted area [Mackay and Cherry, 1989].

Although PAT is generally quite efficient to prevent the contaminant plume from migrating further, it seldom achieves cleanup concentration goals for groundwater. Several authors [Cherry *et al.*, 1990; Kerr, 1987; Armstrong, 1991] reported that the rate of contaminant mass removal from a system (chemical mass per time) decreases when the groundwater pumping rates are increased. They suggested that this phenomenon was the result of the transfer from the NAPL to the groundwater becoming limited in some way. Kerr [1987] suggested that desorption kinetics may act to limit the contaminant removal from the aquifer. The same phenomenon was observed by Armstrong [1991] for vapour extraction systems, where the contaminant removal becomes limited by the partitioning between liquid and vapour phases. Cherry *et al.* [1990] estimated that the rate of contaminant flow would have to be increased by a factor of at least 5 to 10 in order for dissolution of a NAPL to be significantly increased, and therefore flushed away. This could only be achieved close to pumping wells, however, and the radius of influence around such wells is generally quite small. It was therefore their conclusion that it will be difficult to efficiently increase the rate of groundwater flow through NAPL zones, unless the pumping or injection wells are located directly within the NAPL zones. This is often very difficult to achieve, however, and may even make the situation worse if, for example, isolated pools of free product are created (as they will then remain permanent sources of contamination). Well borings themselves can also constitute conduits for contaminants to propagate to lower, perhaps pristine, hydrogeologic units [Gelhar *et al.*, 1992].

To explain these phenomena, affecting the rate of contaminant removal, some authors [*Haley et al.*, 1991; *Mackay and Cherry*, 1989] identified several factors, including (1) hydrogeological factors (heterogeneity, low-permeability units, fractures, dead-end pores, etc.), (2) contaminant-related factors (immiscibility of the contaminant, its affinity for sorption, etc.), (3) continued migration from the source and size of plume, and (4) system design factors (pumping rate, screened intervals, extraction well location, etc.). One of the recommendations made by *Haley et al.* [1991] to improve ground water response actions was the development of methods to enhance extraction effectiveness and efficiency.

In extraction systems pumping at a constant rate, observations show that contaminant concentrations drop significantly initially, followed by a levelling out [*Armstrong*, 1991; *Sundberg*, 1989; *Haley et al.*, 1991]. In cases where continued decreases in concentrations are observed, the concentration decline is usually slower than expected after a period of initial rapid decline [*Haley et al.*, 1991]. As another consequence of sorption, if pumping is stopped before all of the contaminants are removed, the contaminant concentrations in the groundwater will rise as desorption continues to take place. *Sundberg* [1989], at a site contaminated by organic chemicals near Stockton (CA), noted that after a period of high recovery rate, there was a rapid levelling out of the contaminant mass reduction curve. As part of this study, different alternatives were considered to increase the rate of cleanup in terms of mass of contaminant per day removed from the aquifer system. Those alternatives included increasing the pumping rate, relocating the pumping wells, adding new wells, and the conversion of monitoring wells to pumping wells. One of the options was to operate the existing wells in a cyclic fashion, i.e. to turn the pumps on and off to induce flushing of the vadose zone by the rising and falling of the water table. This approach to PAT is often called “pulsed” or “intermittent” pumping. Pulsed pumping was also tested by *Armstrong* [1991] for a vapour extraction system. Both studies concluded that no significant improvement in terms of contaminant recovery were achieved using this technique. Although both authors found pulsed pumping not to be a significant improvement over continuous pumping, their results were based on very few observations. More recently, *Mackay et al.* [2000]

performed an extensive field study conducted at the Dover Air Force Base (Dover, DE, USA) to investigate the impact of pulsed pumping on contaminant recovery. Their study compared the contaminant recovery efficiency in two adjacent cells to isolate two portions of an existing contaminant plume using sheet piling. Continuous pumping was used in the first cell, while pump operations were interrupted intermittently in the second. They concluded that marginal improvements were achieved by intermittent pumping. They also found that while spatial heterogeneity of the aquifer plays an important role on the efficiency of remediation, spatial variations in the initial (pre-remediation) distribution of the contaminant, as well as the nature and extent of the contamination in adjacent areas of low permeability, also influenced the efficiency of the restoration effort.

A new approach to subsurface remediation, *Monitored Natural Attenuation* (MNA), is slowly gaining acceptance as a viable alternative to costly, often inefficient, aquifer cleanup techniques such as excavation or PAT [National Research Council, 1997]. The regulatory definition of “natural attenuation” includes all natural phenomena that contribute to reduce the concentration of a contaminant in water [National Research Council, 2000]. MNA is defined formally by the United States Environmental Protection Agency (USEPA) as including “biodegradation; dispersion; dilution; sorption; volatilization; radioactive decay; and chemical or biological stabilization, transformation, or destruction of contaminants.” [U.S. Environmental Protection Agency, 1999b]. In order to demonstrate that natural attenuation is a valid alternative to the remediation of a contaminate site, the cause-and-effect relationship between the loss of contaminant and the mechanism responsible for the loss must generally be demonstrated [Bekins *et al.*, 2001; National Research Council, 2000]. Dispersion is possibly one of the least predictable of these processes, due in part to the fact that it is highly sensitive to spatial and temporal variability in hydrogeologic conditions and that the inter-dependency between these two sources of variability is poorly understood. Moreover, dispersion is expected to have an impact on the efficiency of the other natural attenuation processes. For example, biodegradation benefits from the reduction-oxidation (redox) conditions generally found near the centre of a contaminant plume. Prediction of the degradation rates is strongly dependent upon the extent of plume spreading due to dispersion.

1.3 Hydrodynamic dispersion

When water flows through granular materials, dissolved solutes are forced along a tortuous path, which causes them to occupy an ever-increasing portion of the aquifer. *Bear* [1972] describes hydrodynamic dispersion as "... the macroscopic outcome of the actual movements of the individual tracer particles through the pores and the various physical and chemical phenomena that take place within the pores." [p. 580]. Unsteady flow fields can also enhance this dispersion by imposing an additional, transient source of variability on the flow field [e.g. *Kinzelbach and Ackerer*, 1986; *Goode and Konikow*, 1990; *Rehfeldt and Gelhar*, 1992].

1.4 Spatial variability

While hydrodynamic dispersion occurs even in the simplest porous media, the tortuous arrangement of pore-scale velocities is even more complex in spatially variable (heterogeneous) aquifers, causing plume dispersion to become even more pronounced than in homogeneous systems. It is generally accepted that spatial variations in the hydraulic conductivity of porous materials largely contributes to the spreading of solutes dissolved in groundwater [*Gelhar et al.*, 1992].

The use of deterministic models to simulate flow and transport in heterogeneous aquifers requires that the input parameters (e.g. hydraulic conductivity, distribution coefficients, porosity, etc.) be specified explicitly at each node in the model's domain. *Ababou et al.* [1989] suggested that the minimum requirement to adequately represent a heterogeneous three-dimensional aquifer is on the order of 10^6 nodes. It is clearly impractical (if not impossible) to make measurements at each of the hundreds of thousands of nodes that would be required. Stochastic theories [*Gelhar and Axness*, 1983; *Dagan*, 1984] circumvent this need for a deterministic characterization by treating the spatial variability of the flow parameters as a random stationary process. These techniques rely on the statistical properties of the medium, such as mean and variance, which also demand a relatively large number of data points, although this number is many times smaller than for deterministic methods.

In order to obtain meaningful estimates of parameters such as hydraulic conductivity (K) or distribution coefficients (K_d), an extensive sampling network is required, resulting in the need for a tremendous number of sampling points. The investments, both in terms of money and time, required for detailed, three-dimensional sampling studies are very high, and few such investigations exist. The better known high-resolution geostatistical analyses of the spatial distribution of K from field experiments include those performed at sites located at CFB Borden, Ontario [Sudicky, 1986], Cape Cod, Massachusetts [Hess *et al.*, 1992], and Columbus, Mississippi [Rehfeldt *et al.*, 1992]. Table 1-1 summarizes the results from those studies and clearly shows that the spatial distribution of K at these sites is highly variable.

Table 1-1. Summary of selected detailed geostatistical investigations of the spatial variability of hydraulic conductivity, K

	Borden, ON	Cape Cod, MA	Columbus, MS
	<i>Sudicky [1986]</i>	<i>Hess et al. [1992]</i>	<i>Rehfeldt et al. [1992]</i>
Analysis method	laboratory permeameter	laboratory permeameter	borehole flowmeter
Number of coring locations	32	16	16
Number of samples, N	1279	825	668
Vertical spacing, m	0.05	0.15	0.073
Horizontal spacing, m	1.0	0.9-24.0	0.9-24.0
Geometric mean, K_g , cm/s	0.00975	0.11	0.035
Variance of $\ln K$, $\sigma^2_{\ln K}$	0.38	0.24	0.14
Minimum value of K , cm/s	0.0006	0.013	0.006
Maximum value of K , cm/s	0.02	0.37	0.14

1.5 Temporal variability

Most field and numerical studies assume steady state groundwater flow. This assumption is typically justified by considering the temporal flow fluctuations to be negligible. However, this decision of using a steady state model is more commonly made due to the lack of medium- to long-term data on the flow system. A number of natural and artificial stresses can cause fluctuations in flow direction and/or magnitude. In fact, at those sites where the collection of water-level data covers a relatively long period, conditions usually show evidence of natural transience. This suggests that unsteady flow conditions

may be the norm rather than the exception in most natural settings. This is especially true in climates where the water table is often shallow, and where regional gradients are controlled largely by net infiltration rates. In these areas, the amount of recharge can be important, but distributed in an uneven fashion throughout the year. In Canada and the northern United States, for example, infiltration is much higher in spring compared to winter due to the melting snow and spring rain infiltrating through the thawing ground; infiltration slows down during the hot summer months due to increased evapotranspiration, but becomes higher in fall when the vegetation stops photosynthesizing. It is also common to observe fluctuations in precipitation rates over periods of several years. The effect of such changes in recharge may greatly affect the water table configuration and groundwater flow regime. *Freeze and Cherry* [1979] listed some of the natural and man-induced mechanisms that cause fluctuations in groundwater levels. These are summarized in Table 1-2, along with some of their characteristics.

Table 1-2. Summary of natural and artificial mechanisms that may cause fluctuations in groundwater levels [from *Freeze and Cherry*, 1979 (p. 230)]

Mechanism	Characteristics							
	Unconfined	Confined	Natural	Man-Induced	Short-lived	Diurnal	Seasonal	Long-term Climatic Influence
Groundwater recharge (infiltration to the water table)	✓		✓				✓	✓
Air entrapment during groundwater recharge	✓		✓		✓			✓
Evapotranspiration and phreatophytic consumption	✓		✓			✓		✓
Bank storage effects near streams	✓		✓				✓	✓
Tidal effects near oceans	✓	✓	✓			✓		
Atmospheric pressure effects	✓	✓	✓			✓		✓
External loading of confined aquifers		✓		✓	✓			
Earthquakes		✓	✓		✓			
Groundwater pumping	✓	✓		✓				✓
Deep-well injection	✓	✓		✓				✓
Artificial recharge: leakage from ponds, lagoons, landfills	✓			✓				✓
Agricultural irrigation and drainage	✓			✓				✓
Geotechnical drainage of open pit mines, slopes, tunnels, etc.	✓			✓				✓

1.6 Dispersion under transient flow: previous work

It is generally accepted that spatial heterogeneities in porous formations play a major role in the dispersion of contaminants in groundwater. This is thought to be the result of solutes following preferred trajectories along the zones of higher hydraulic conductivity. Recently, many authors have suggested through theoretical work that variations in the magnitude and direction of groundwater flow fields may also enhance solute dispersion. Such variations, since they vary in time, are referred to as transient processes. In order to account for such flow transients, a deterministic approach was adopted by some [e.g. *Kinzelbach and Ackerer*, 1986; *Goode and Konikow*, 1990; *Naff et al.*, 1989], while

others addressed the issue of transience in a stochastic framework [Rehfeldt and Gelhar, 1992; Rubin and Bellin, 1994; Zhang and Neuman, 1996; Dagan et al., 1996; Bellin et al., 1996].

Table 1-3 summarizes some of the conclusions from these studies, specifically on the effects of variations in the magnitude and/or direction of hydraulic gradients on solute dispersion. Most of these studies were concerned with the influence of gradient direction variations on macrodispersion; some also investigated the effects of unsteady gradient magnitude. Conclusions suggest that variations in the flow direction either have a negligible impact on, or slightly reduce, longitudinal dispersion (A_x). Transverse dispersion (A_y), on the other hand, seems to be invariably enhanced by flow direction transients. When they were incorporated into the analysis, gradient magnitude variations were shown to increase A_x , while the effect on A_y was found to be small, increasing in most cases, but sometimes behaving in an inconsistent pattern.

Table 1-3. Summary of the theoretical predictions of the influence of transient flow fields on solute dispersion in porous media. The plus (+) and minus (–) signs indicate an increase or decrease in predicted dispersivity, respectively, while the symbol +/- is used where a significant effect was observed, but where the trend was unclear. The symbol ≈ 0 is used where a negligible effect was reported. Empty cells indicate that the corresponding quantity was not addressed by the study.

Reference	Direction variations		Magnitude variations	
	A_x	A_y	A_x	A_y
Kinzelbach and Ackerer [1986]	–	+		
Goode and Konikow [1990]	– ($\alpha_T/\alpha_L < 0.5$)	+		
	+ ($\alpha_T/\alpha_L > 0.5$)			
Naff et al. [1989]	≈ 0	+		
Rehfeldt and Gelhar [1992]	≈ 0	+	+	+
Adams and Gelhar [1992]			+	+/-
Rubin and Bellin [1994]			+	+
Zhang and Neuman [1996]	–	+	+	+/-
Dagan et al. [1996]	≈ 0	+		
Bellin et al. [1996]	≈ 0	+		

Using a two-dimensional random-walk transport model, Kinzelbach and Ackerer [1986] performed numerical simulations that showed the apparent transverse dispersivity (α_{Ta}) to

be measurably larger under time-varying flow conditions than under steady-state flow. Apparent longitudinal dispersivity (α_{La}), however, was found to be comparatively smaller, since $\alpha_{La} + \alpha_{Ta} = \alpha_L + \alpha_T$, where α_L and α_T represent the true longitudinal and transverse dispersivity values, respectively.

Goode and Konikow [1990], also using a deterministic approach, suggested that α_L is increased by transient flow fields, and that the changes in the direction of flow have a greater effect on dispersion than changes in flow magnitude. Their study also suggested that asymmetric transients, similar to those induced by pumping, may significantly increase α_T . Contrary to *Kinzelbach and Ackerer* [1986], transient flow fields were shown to increase α_L , especially for ratios α_T/α_L close to unity. Such a high ratio is unrealistic under most field conditions, however, where groundwater velocity is generally high enough to render molecular diffusion negligible. In such advection-dominated systems, the ratio α_T/α_L is generally much smaller [*Freeze and Cherry*, 1979]. In this context, *Goode and Konikow* [1990] found that variations in the hydraulic gradient direction caused α_L to decrease, in accordance with the findings of *Kinzelbach and Ackerer* [1986].

Sudicky [1986], using data collected during the 1982-85 Stanford-Waterloo slug tracer experiment at the CFB Borden site, estimated the macrodispersivity of a conservative tracer cloud. These dispersivity estimates, as predicted by previously published stochastic theories [*Gelhar and Axness*, 1983; *Dagan*, 1982; *Dagan*, 1984], were compared to the macrodispersivities derived from field data, and calculated from spatial moments by *Freyberg* [1986]. This comparison showed that both models were capable of adequately predicting longitudinal and vertical transverse dispersion, but significantly under-predicted the horizontal transverse component of macrodispersivity. Since groundwater flow transients are not taken into account by either of these models, *Sudicky* [1986] suggested that the transience in the hydraulic gradients might explain the unusually large data-derived transverse macrodispersivity.

In order to account for the large lateral dispersion observed at the CFB Borden site by *Freyberg* [1986], *Naff et al.* [1989] incorporated the effects of the horizontal transverse flow transients into their estimation of the second spatial moments of solute plume.

According to this model, a series of harmonic functions, determined from a spectral analysis of the water level history, can be summed in order to characterize flow transients. Spatial variability in hydraulic conductivity heterogeneity is not taken into account by this deterministic model, which assumes temporal variability in the flow field to be the principal contributor to solute spreading. Using this original technique, *Naff et al.* [1989] thus successfully predicted horizontal transverse moments which compared favourably with field observations.

In a theoretical study, *Rehfeldt and Gelhar* [1992] extended the stochastic continuum theory of *Gelhar and Axness* [1983] to include a transient term, and derived a numerical expression for the asymptotic macrodispersivity tensor in three-dimensional, heterogeneous porous media. This tensor was shown to be the sum of (a) the spatial variability (also referred to as the input covariance describing the hydraulic conductivity), and (b) the temporal variability (or input covariance of the hydraulic gradient). Their study assumed that temporal variability was independent from spatial variations in hydraulic conductivity, while allowing for both sources of variations to contribute to solute cloud dispersion. They suggested that variations in hydraulic gradient magnitude enhance longitudinal dispersivity, whereas varying the gradient direction increases transverse dispersivity. In order to incorporate flow transience into their analysis, only a limited amount of information about the fluctuating groundwater elevations at the CFB Borden site was available to *Naff et al.* [1989] and *Rehfeldt and Gelhar* [1992]. Some of the input model parameters they used were therefore selected subjectively in the application of their respective model to field data. Nevertheless, their results yielded improved estimates of transverse horizontal macrodispersivity, compared to previous theories that neglected flow transience altogether.

More recently, a second field tracer experiment involving the migration of an emplaced DNAPL contaminant source under natural gradient conditions took place (1989-91) at the CFB Borden test site, 150 m away from the original test [*Rivett et al.*, 1994c]. Throughout the duration of this test, a detailed record was kept of the groundwater elevations across the facility in order to capture the flow transients. The direction of the regional hydraulic gradient varied by 30 degrees during the test; gradient

magnitude fluctuated between 0.33% and 0.64% across the site. The temporal evolution of the water table was explicitly accounted for in numerical simulations performed by *Rivett et al.* [1994b] aimed at comparing transport parameters from the two adjacent field sites. They showed that even though they were extremely close to one another, significant differences existed between the test sites. In particular, horizontal transverse dispersivity and solute retardation factors were both sizably smaller than they were during the original experiment.

Farrell et al. [1994b] also used transient water level data collected during the 1989-91 test to verify how closely horizontal transverse dispersion could be predicted by the models of *Rehfeldt and Gelhar* [1992] and *Naff et al.* [1989]. In the first case, results proved to satisfactorily forecast macrodispersion at the site. The second model significantly under predicted the measured dispersivity. *Farrell et al.* [1994b] pointed out, as did *Rivett et al.* [1994b], that "... observed gradient variations in time are actually quite small at the Borden site, yet they account for almost all of the horizontal spreading."

Dagan et al. [1996], combining deterministic transients with a spatially random $\ln(K)$ distribution, investigated the effects on dispersion of non-steady transients in the hydraulic gradient direction. They suggested that periodic fluctuations in the direction of flow had an insignificant effect on longitudinal dispersion, while strongly increasing transverse dispersion by adding a constant source of dispersivity. They found an interaction to exist between the spatial and temporal variability. Applying this model to the CFB Borden data, *Bellin et al.* [1996] attributed the enhanced transverse dispersion to spatial heterogeneity, combined with the effect of the time-variations in the hydraulic gradient. Flow transients, on the other hand, were found to have a minor impact on the longitudinal component of dispersion compared to steady state conditions.

Rubin and Bellin [1994], through a nonlinear transformation of the travel time scale derived a space random function describing groundwater velocity in a heterogeneous, uniformly recharged aquifer. Using this method, their study investigated the case of non-uniform, unidirectional flow, using the recharge to induce transients in the gradient magnitude. Their theoretical results comparing transient to steady state flow suggested

that longitudinal and transverse dispersion are both enhanced by recharge, although this effect was more significant in the former. Similarly, in an attempt to explain the larger-than-predicted longitudinal plume spreading observed at the Columbus site, *Adams and Gelhar* [1992] suggested that the accelerating nature of the flow magnitude had to be taken into account. They found that the effect of the non-uniform flow was to increase the longitudinal variance significantly, while possibly decreasing lateral (transverse) dispersion.

Zhang and Neuman [1996] used an original approach to investigate the effects on dispersion of variations of both gradient magnitude and direction. They used low-order approximations of the autocovariance and cross-covariance of velocity, head, and $\ln(K)$ under quasi-steady state flow conditions, where flow adjusts to changes in boundary conditions instantly, generating scenarios consisting of a series of steady states. Contrary to the findings of *Rehfeldt and Gelhar* [1992], their results showed that gradient magnitude variations increase longitudinal dispersion while transverse dispersion oscillates around the steady state value. Conversely, variations in the direction of the gradient cause longitudinal dispersion to decrease and transverse dispersion to increase, in agreement with *Kinzelbach and Ackerer* [1986] and *Goode and Konikow* [1990].

2 OBJECTIVES AND SIGNIFICANCE

The principal objective of this work is to characterize the effects of transient groundwater flow on contaminant migration in porous geologic materials. More specifically, to assess quantitatively the dispersion of miscible solutes subjected to unsteady flow fields, through a series of laboratory experiments and numerical simulations. The hypothesis to be tested is that the nature and the form of flow transience have a great influence on the pathways taken by dissolved contaminants. This study will contribute to the current state of understanding of the dispersion of dissolved substances in groundwater, in particular when the flow field is subjected to time-dependent (transient) variations. This work will demonstrate, by conducting in parallel laboratory experiments and numerical simulations, that the theory does not always accurately predict the movement of dissolved contaminants, particularly when complexity in hydrogeologic conditions (in space or in

time) increases. This is achieved using an innovative physical model that provides the means to study the behaviour of a spreading tracer plume evolving through homogeneous or heterogeneous porous media that can be subjected to deterministic flow transients.

More specifically, we will address the following questions:

- 1. Is it possible to reproduce in the laboratory the complex hydraulic conditions encountered in nature, in particular by combining flow transients with heterogeneous porous media ?**

The innovative design of the laboratory model, consisting of a two-dimensional flow cell and coupled hydraulic control system, allowed us to construct spatially homogeneous or heterogeneous porous media of prescribed statistical properties, and to impose deterministic flow transients on the system.

- 2. Is it possible to accurately monitor using a physical model the spatial distribution and temporal evolution of a contaminant plume migrating through homogeneous or heterogeneous porous media ?**

A monitoring procedure that combines image processing with spatial moment analysis is presented. These tools can characterize with great spatial and temporal resolution the evolution of contaminant plumes, as measured from sequences of digital images acquired during the course of laboratory experiments.

- 3. What are the impacts on macroscopic dispersion of transience in the hydraulic gradient, both in terms of its magnitude and its direction, and how does this compare with previously published results ?**

A series of flow scenarios, each including identical initial and boundary conditions, will be applied to homogeneous and heterogeneous systems using the laboratory and numerical models in parallel. The impact of variations in the flow field on solute plume dispersion will be assessed by monitoring the dispersive behaviour of contaminant plumes as they are subjected to transient gradients. We will show that the influence of flow transience on solute dispersion, as characterized in this study, compares well with similar results reported in the literature, based on theoretical or numerical investigations.

4. Do flow transients influence contaminant dispersion differently in heterogeneous compared to homogeneous porous media ?

Although it is clear that spatial variability (heterogeneity) increases solute spreading, and that temporal variability (flow transience) enhances solute spreading, it is still not clear whether these factors are independent of each other.

5. Can the commonly used numerical models MODFLOW and MT3D reproduce the experimental results obtained during laboratory experiments under controlled steady and transient flow conditions ?

A comparison of the experimental results from numerical models to those of the corresponding laboratory experiments will show that while the fit is generally good for simple systems involving a homogeneous porous medium subjected to steady flow conditions, the increased complexity introduced by the spatial and temporal variability can lead to inconsistencies between the computer model and the laboratory model.

CHAPTER 2 – Methodology

“There are three principal means of acquiring knowledge... observation of nature, reflection, and experimentation. Observation collects facts; reflection combines them; experimentation verifies the result of that combination.”

– Denis Diderot (1713 - 1784)

The tools and techniques that were used in this work are described summarily in this chapter. The reader is referred to the appendices for the corresponding detailed descriptions, including (A) *Laboratory Apparatus*; (B) *Image Processing*; (C) *Spatial Moments*; (D) *Macrodispersivity Calculation*; (E) *Reynolds Number*; and (F) *Wall Effects*.

This investigation involved the design of a laboratory model (flow cell) and accompanying numerical model. The physical and numerical models are described in the following section. To monitor the dissolved plume migrating in the flow cell, image sequences were acquired and processed to produce concentration maps (see § 1.4). The analysis of the laboratory and numerical results was performed by tracking the temporal evolution of the spatial moments derived from these datasets (see § 3). Ultimately, the macroscopic dispersive behaviour of the contaminant plumes was characterized by calculating the macrodispersivity from the normalized first- and second-order spatial moments (see § 4).

1 EXPERIMENTAL DESIGN

Vital to the completion of this study was the construction of a physical model capable of reproducing – at a smaller scale – realistic field conditions of groundwater flow. In particular, the main challenge was to design a flow tank and associated hydraulic control

apparatus that allowed uniform flow fields to be easily and rapidly modified “on the fly”. These temporal changes in the mean flow conditions (flow transience) had to be imposed on the system while preserving equipotential lines parallel to one another.

Appendix A provides a detailed description of the physical characteristics of the flow tank used in this study, as well as the various components used to establish groundwater flow and dynamically vary hydraulic conditions in the flow tank.

1.1 Homogeneous porous media

The experiments were organized in a factorial design to assess the effects on solute dispersion of the two principal aspects of the hydraulic gradient, that is, its magnitude and its direction. The word “direction” is used loosely here, to describe both spatial and temporal changes in the hydraulic gradient. Two values (levels) of gradient magnitude ($i = dh / dx = 0.12$ and 0.22) were combined with four levels of gradient direction (steady state, shifting direction with a low (30°) or a high (60°) rotation angle, and reversing). As a result, eight flow scenarios were investigated, as summarized in Table 2-1.

Table 2-1. Summary of experimental and numerical flow scenarios (cases). The number of replicates performed during the laboratory experiments (η) is indicated for each case. The two principal factors (gradient magnitude and direction) are indicated in capital letters. Eight flow configurations, or cases, were considered for the homogeneous porous medium (labelled 1 through 8). For the heterogeneous work, a single value of the hydraulic gradient was used: those cases are labelled 9 through 12.

Hydraulic Gradient, i	Configuration of Hydraulic Gradient Direction			
	Steady State	Shifting (“zigzag”)		Reversing
		$\pm 30^\circ$	$\pm 60^\circ$	
0.12	<u>Case 1</u> ($\eta = 6$)	<u>Case 2</u> ($\eta = 3$) $\Delta t_1 = 5; \Delta t_2 = 10$	<u>Case 3</u> ($\eta = 2$) $\Delta t_1 = 5; \Delta t_2 = 10$	<u>Case 4</u> ($\eta = 2$) $\Delta t_1 = 10; \Delta t_2 = 5$
0.22	<u>Case 5</u> ($\eta = 2$)	<u>Case 6</u> ($\eta = 2$) $\Delta t_1 = 2.5; \Delta t_2 = 5$	<u>Case 7</u> ($\eta = 2$) $\Delta t_1 = 2.5; \Delta t_2 = 5$	<u>Case 8</u> ($\eta = 2$) $\Delta t_1 = 5; \Delta t_2 = 2.5$
0.21	<u>Case 9</u> ($\eta = 1$)	<u>Case 10</u> ($\eta = 1$) $\Delta t_1 = 2.5; \Delta t_2 = 5$	<u>Case 11</u> ($\eta = 1$) $\Delta t_1 = 2.5; \Delta t_2 = 5$	<u>Case 12</u> ($\eta = 1$) $\Delta t_1 = 5; \Delta t_2 = 2.5$

a. Gradient direction variations occurred at time intervals indicated (in minutes) by Δt_1 (first and last periods) and Δt_2 (all other periods in between). Gradient direction variations for “Shifting” cases expressed relative to the positive longitudinal (x) direction.

Model Description

This section provides a brief summary of the laboratory and numerical models. The experimental setup is presented in detail in Appendix A, including a description of (1) the physical model (flow cell and hydraulic control system), (2) the physical properties of the porous medium and emplacement (packing) methods used, (3) the physical and optical characteristics of the tracer (dye), and (4) the image acquisition system. The numerical model used to replicate the experimental conditions observed in the laboratory is summarized below.

Laboratory model

The laboratory experiments in this study were carried out using a two-dimensional flow cell and associated hydraulic control system that were designed specifically for this study. The flow cell was used to see and quantify the advection and dispersion of a tracer cloud transported by flowing groundwater. The flow cell and the granular porous medium it contained were translucent, enabling direct observation of the tracer plume within the cell when light was transmitted through the cell from behind. The light transmission technique is an adaptation of a similar design presented by *Tidwell and Glass* [1994]. The innovative circular design of the flow cell allowed for variations in the direction of the uniform gradient direction with little interference from the flow cell geometry. The hydraulic control system (HCS) consisted of 12 constant head input/output reservoirs connected to 12 ports around the flow cell, via a 12-channel hydraulic switch. This 12-channel arrangement provided rapid changes in the direction and magnitude of the uniform gradient.

For the experiments reported in Chapter 3, the flow cell was uniformly packed with a homogeneous porous medium composed of well sorted, spherical glass beads, with mean diameter $\phi = 335 \mu\text{m}$, porosity $n = 0.38$, and hydraulic conductivity $K = 0.103 \text{ cm s}^{-1}$ (see Appendix A for a complete description of the physical properties and packing method). The eight flow scenarios presented were run using the same packing. Each scenario was run between two and six times (as indicated by η in Table 2-1).

Prior to each experiment, the initial flow regime was established and the system was allowed to equilibrate and reach steady state flow conditions. To verify this, 12 manometers – one per channel – were monitored until hydraulic heads variations in each channel were within ± 1 mm.

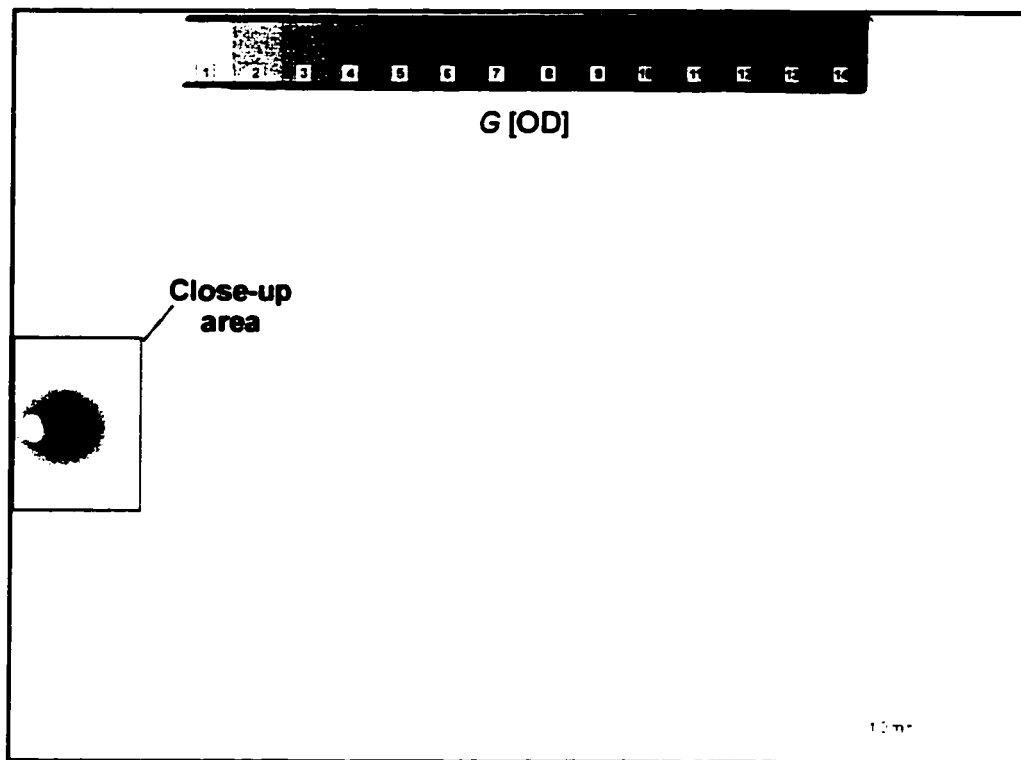
Once steady state conditions were reached, a tracer pulse was introduced into the model. Flow in the cell was stopped temporarily in order to minimize smearing and to obtain a reproducible initial pulse. A 10 mL slug of 8.40 mg L^{-1} Rhodamine WT dye (RWT) was slowly injected through the injection port. Introduction of the tracer into the model took place over a period of approximately 30 s, in exactly the same manner for all experiments. The initial shape of the diluted plume was cylindrical, occupying the full thickness of the flow cell (0.033 m), and centred at the injection port, located near the upstream end of the flow cell. A stopwatch was turned on at exactly the same time flow was allowed to resume within the flow cell, marking the starting time, $t = 0$. In order to monitor the spatial and temporal evolution of the solute plumes, a series of greyscale digital images was acquired for each experiment at a regular interval of 30 or 60 seconds depending on the case.

Numerical model

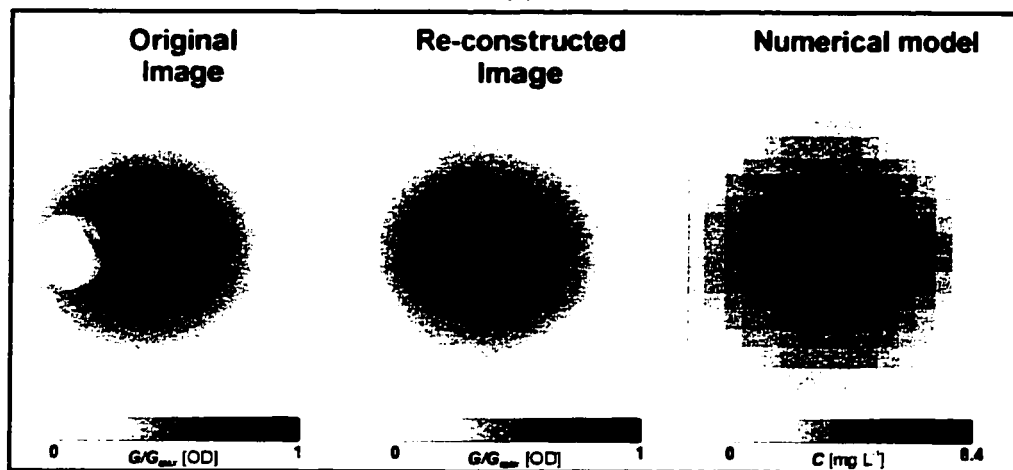
A numerical model was used to simulate as closely as possible the experimental conditions from the laboratory model. The 0.65 m wide by 0.65 m tall (x and y directions, respectively) finite difference grid was generated using 172 columns and 172 rows, resulting in 29584 square cells with a width and height equal to 0.00379 m (3.79 mm). Because of the two-dimensional design of the flow cell, velocities in the third dimension (z direction) were expected to be negligible. Moreover, because solute plumes occupied the full thickness (0.033 m) of the flow cell, vertical concentration gradients were also insignificant. For these reasons, a single layer was used in this three-dimensional numerical model to approximate the two-dimensional flow cell described in the previous section. The homogeneous system properties were selected to match those of the physical model, including an isotropic hydraulic conductivity $K_x = K_y = K_z = 0.103 \text{ cm s}^{-1}$ and a

uniform porosity $n = 0.38$. Because molecular diffusion in such permeable materials is expected to be negligible, a very small value of $D_m = 10^{-10} \text{ m}^2 \text{ s}^{-1}$ was used.

Initial concentrations (C_0) near the injection port were estimated using an image from one of the laboratory experiments, and were set to zero elsewhere in the flow domain. The solute distribution from a steady state experimental run (case 1, run 1) was measured from the image acquired at $t = 1 \text{ min}$ (shown in Figure 2-1(a)). A close-up of the area located near the injection zone is shown on the left in Figure 2-1(b). Notice how the injection port, visible as a light-coloured object, hides a portion of the plume. In order to “remove” this opaque object from the image, the right half of the plume was mirrored vertically using the column of pixels corresponding to the centre of mass as the pivot. The middle image in Figure 2-1(b) shows this reconstructed image. Finally, pixel values from the image (each $0.47 \times 0.47 \text{ mm}^2$) were averaged and assigned to the coarser cells ($3.79 \times 3.79 \text{ mm}^2$) of corresponding position on the finite difference grid. Each grid cell in the numerical model is therefore representative of approximately 64 (i.e. 8×8) pixels from the image. The resulting initial concentration map shown on the right of Figure 2-1(b) was used as the initial condition for all transport simulations discussed in this chapter.



(a)



(b)

Figure 2-1. (a) Image from a laboratory experiment (case 1, run 1) acquired 1 min after tracer injection. This image was used to generate the initial concentration values assigned in each grid cell in the numerical model, used in all simulations. Pixel values were expressed as G/G_{MAX} , where G is the standardized optical density (OD) at a given pixel, and $G_{MAX} = 10.3$ OD is the image maximum. The area outlined in (a) is enlarged and shown in (b), along with the image (centre) reconstructed by mirroring the right half of the plume to estimate the solute distribution hidden behind the injection port (visible as a bright object in the image on the left). The resulting map showing the relative initial concentrations (C/C_0) used in the numerical models is shown on the right, with values ranging from 0 (white) to 8.4 mg L^{-1} (darkest).

The intrinsic dispersion coefficient α , or simply “dispersivity”, is an input parameter used by numerical models to account for mixing processes that occur at spatial scales smaller than the representative elementary volume (REV). The advection-dispersion equation (ADE), as its name implies, describes solute transport in groundwater, through an advective term and a dispersive term. The dispersive effects due to the complex nature of the velocity field are unaccounted for in the advective part of the ADE, which treats the porous medium as a continuum entirely open to flow [de Marsily, 1986]. Small-scale dispersivity values were adjusted by calibration using the experimental data obtained from a simple flow scenario (steady state case 1, run 1). Longitudinal and transverse dispersivity (α_L and α_T , respectively) were adjusted until the extent of the simulated solute plume matched that of the experimental data for that scenario, at $t = 5, 10$ and 25 minutes. The calibrated estimates ($\alpha_L = 0.00045$ m and $\alpha_T = 0.0000225$ m) were then used as input values in the numerical simulations for all flow scenarios (cases 1 through 8). The ratio $\alpha_L/\alpha_T = 0.05$ falls within the typical range of values, which is generally between 0.2 and 0.01 [de Marsily, 1986].

Hydraulic heads measured in each channel during the course of the laboratory experiments were used as input in the numerical simulations.

1.2 Heterogeneous porous media

The approach used here is almost identical to that of the homogeneous cases as described in the preceding section, the only differences being the non-uniform hydraulic properties of the porous medium, and the fact that only one magnitude of the hydraulic gradient was used. The same steady and transient flow patterns were applied to the flow cell as for homogeneous porous medium cases; however, the porous medium was replaced by a stochastically derived heterogeneous medium. The four flow scenarios were: steady state, zigzag (with rotation angles of 30° and 60°) and reversing flow.

This study used a single realization of a randomly generated hydraulic conductivity field with prescribed statistical properties. This random field was used to build both the laboratory and numerical models. Incorporating complex hydrogeologic structures, which

may be derived from field data (e.g. using interpolation or kriging) or by randomly generating a synthetic aquifer (as in this study), is not a difficult task when building a numerical model. In the laboratory, on the other hand, building spatial heterogeneities into a physical model is far from trivial because of the challenges facing the experimenter when attempting to accurately reconstruct a specific arrangement of hydraulic properties. Fortunately, the laboratory model's "front loading" design allowed for bi-dimensional structures varying in complexity to be easily (albeit tediously) constructed.

The characteristics of the stochastically derived random field are described below, followed by a description of the methods used to incorporate this synthetic aquifer into the laboratory and numerical models.

Random field generation

A heterogeneous spatial arrangement of log-normally distributed hydraulic conductivity values was randomly generated from prescribed statistical properties using the technique of *Robin et al.* [1993]. An exponential covariance model was selected, with an input mean $\ln(K)$ of -2.74 with K in units of cm s^{-1} and a variance of 0.40 . These properties were selected to approximately match the range of values available from glass beads of nine size classes. The correlation scales ($\lambda_{\ln K}$) were carefully chosen to simultaneously satisfy the lower and upper bounds suggested by *Welty and Elsner* [1997]. First, $\lambda_{\ln K}$ should be at least $1/10^{\text{th}}$ the size of the flow cell, in order for the mean and variance from a single realization to match the values expected from many realizations of the same field. Second, the $\lambda_{\ln K}$ must be large enough to include a minimum of four nodes per correlation scale. These constraints impose the following limits on $\lambda_{\ln K}$:

$$4\Delta x < \lambda_{\ln K} < (0.001 - 0.1)L \quad (2.1)$$

where Δx represents the distance between measurement points and L is the total length of the experiment (domain size). Although the spatial resolution of the images was on the order of 0.05 cm , Δx was interpreted as the dimensions of the blocks used to build the heterogeneous medium ($\Delta x = 1.51 \text{ cm}$). Using a correlation scale of 6.0 cm in both

directions produced a statistically isotropic field that met the lower and upper bounds imposed by (2.1). Using $L = 65$ cm and $\Delta x = 1.51$ cm, we obtain:

$$\lambda_{lnK} = 6.0 \text{ cm} = 4.0 \Delta x$$

$$\lambda_{lnK} = 6.0 \text{ cm} = 0.09 L$$

Using the same input statistics (mean, variance and correlation scales) but changing the initial random number generator seed (an integer number selected at random) produced one out of an infinite number of possible realizations representing the same system. Because of the random nature of the generated fields, the output properties of each realization of the field differ somewhat from the input. The isotropic random field (“ISO-4”) selected in this study showed output statistics that closely matched those specified as input. These are compared in Table 2-2.

Table 2-2. Input and output averages and variances as calculated by the *FGEN* random field generator (version 9.2) with a seed number equal to 111. Figure 2-2 illustrates the relationship between the truncated (a), extracted (b) and binned (c) fields.

ISO-4 Field (seed=111)	Number of cells	Dimensions (cm)		Average In[K (cm s ⁻¹)]	Variance
		Cells	Field		
Input :	1024 × 1024	0.151	154.8 × 154.8	-2.74	0.40
Output :					
Truncated	700 × 700	0.151	105.5 × 105.5	-2.67	0.45
Extracted	430 × 430	0.151	65.0 × 65.0	-2.66	0.52
Final (binned)	43 × 43	1.51	65.0 × 65.0	-2.62	0.37

The full field was created to be deliberately larger and with a finer spatial resolution than the final dimensions needed to match the required $L = 65$ cm and $\Delta x = 1.51$ cm. The procedure used to generate the final (binned) heterogeneous domain from the numerically generated random (full) field is illustrated in Figure 2-2. Starting with an isotropic spatial resolution of 0.151 cm, the full field (1024 × 1024 cells) was truncated to 700 × 700 to eliminate banding problems associated with the spectral method used to generate the random field [Robin *et al.*, 1993]. A 65 × 65 cm² subset whose origin was chosen at random within the (larger) truncated field was then extracted using the same spatial resolution. In order to match the size restrictions imposed by the matrix (template) used

to reproduce this field in the laboratory model, $\ln K$ values from the extracted field were locally averaged in groups of 10×10 (100 cells). Finally, the averaged $\ln K$ values in the 43×43 block domain were assigned one of the nine classes of beads shown in Table 2-3 based on the value of each block. Figure 2-3 shows the distribution of the nine bead classes, numbered 1-9 in decreasing order of permeability.

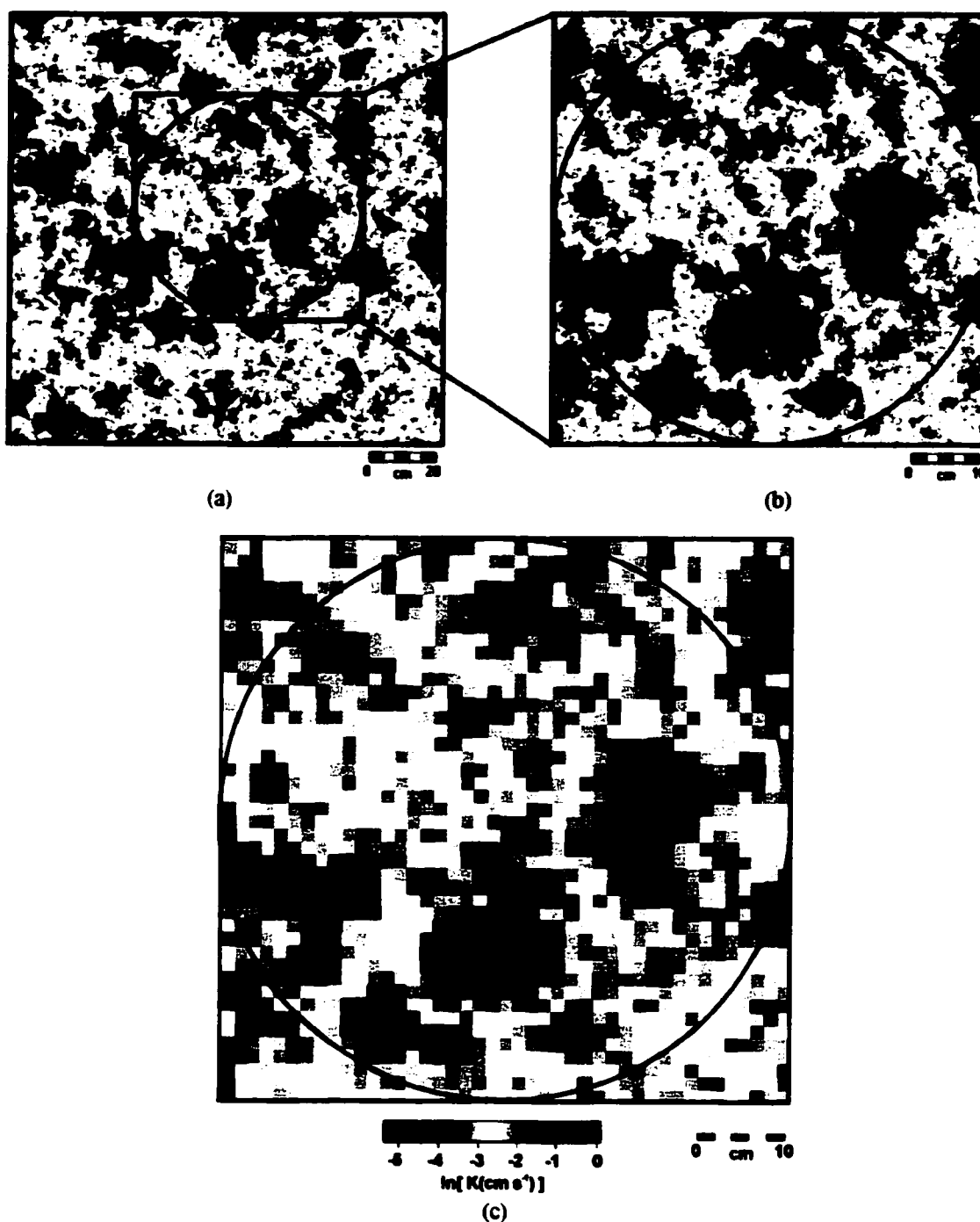


Figure 2-2. Distribution of the hydraulic conductivity values ($\ln K$) produced by *FGEN*. The truncated field (a) was harvested at random to extract the $65 \times 65 \text{ cm}^2$ subset (b). The scale was then reduced by averaging groups of 10×10 cells to produce the final field (c) made of 43×43 averaged square blocks, each 1.51 cm in size (from 0.151 cm cells).

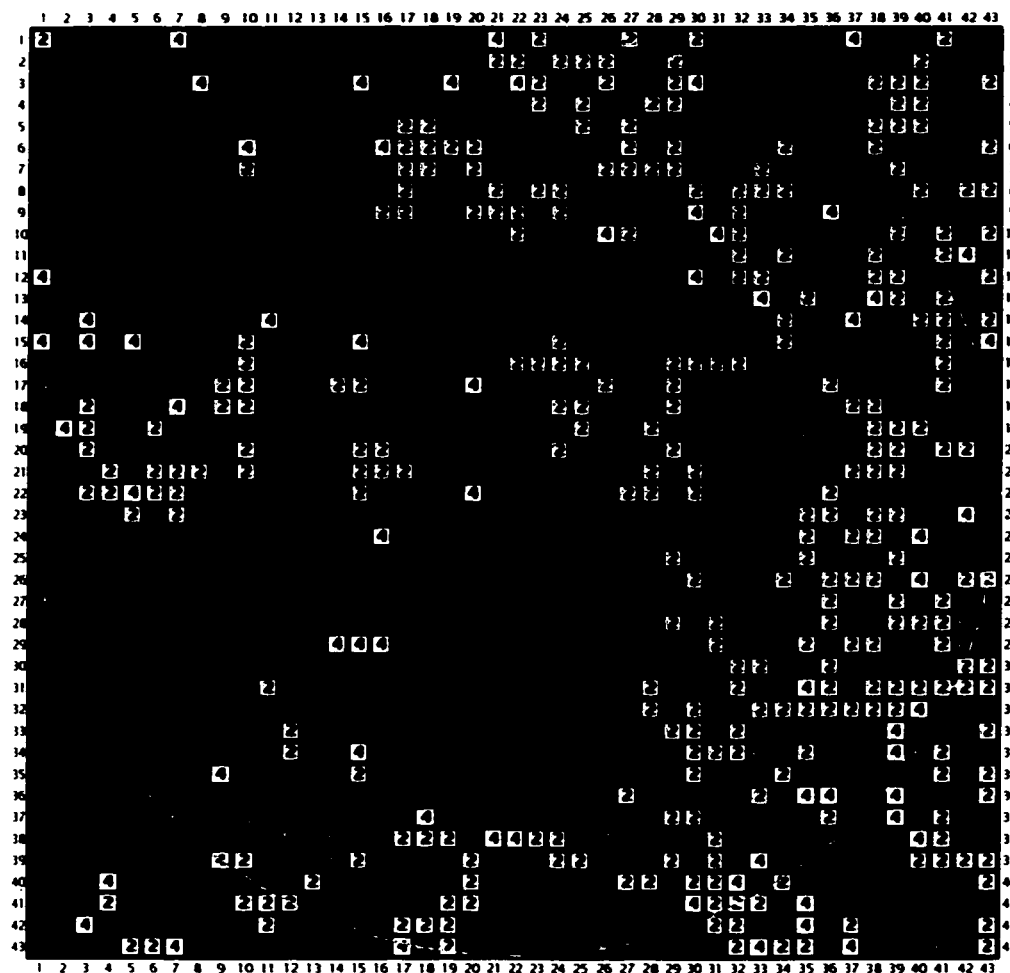


Figure 2-3. Map of the log-normally distributed hydraulic conductivity values ($\ln[K]$) used to construct the heterogeneous porous medium. This distribution was randomly generated on a finer grid (430×430), and then reduced to 43×43 by averaging 100 $\ln[K]$ values in each cell (see Appendix A for details about the random field generation). Each block was assigned a bead class (numbered 1-9) based on its averaged $\ln[K]$ according to Table 2-3. The outline of the circular flow domain is shown in yellow.

Table 2-3. Beads size classes used in Figure 2-3. $\ln K$ ranges for each class are from Figure 2-2(c). K was estimated experimentally using a constant head permeameter setup (see Appendix A for further details).

	Beads	In K range		Bin	count	K cm/s
		From	to			
1	B	-1.9	-1.9	-1.72	228	0.182
2	B+C	-1.9	-2.28	-2.09	229	0.124
3	C	-2.28	-2.51	-2.48	185	0.105
4	C+AA	-2.51	-2.59	-2.55	46	0.078
5	AA	-2.59	-2.79	-2.64	157	0.072
6	AA+AB	-2.79	-3.02	-2.94	160	0.053
7	AB	-3.02	-3.3	-3.1	160	0.045
8	AB+AC	-3.3	-3.58	-3.49	115	0.030
9	AC	-3.58	-3.58	-3.67	148	0.026

Model Description

The two-dimensional heterogeneous permeability structure described in the previous section was used in the construction of the laboratory and numerical models. The two models are in every way identical to those described in the previous section, with the exception that the heterogeneous porous medium was used instead of the homogeneous system presented before. The next sections describe how the porous medium was emplaced within the physical model, and how the numerical model was modified to incorporate the stochastically derived permeability field.

Laboratory model

Emplacement of the heterogeneous porous medium inside the flow cell was done with the help of a template temporarily inserted in the physical model. This template – a thin-walled lattice with 1.5-cm square openings – was machined to fit the circular shape of the flow cell, with a final diameter $\phi = 65.0$ cm and a thickness of approximately 5 cm. The porous medium was assembled one block at a time by pouring granular materials of various sizes (and permeabilities) into the square compartments of the template. The random permeability structure shown in Figure 2-3 was used as a map to fill each of the template's blocks. Figure 2-4 provides an overview of the porous medium emplacement method. Refer to Appendix A for more information related to the construction of the heterogeneous porous medium and details on other components of the experimental setup.

Note that an attempt to minimize the wall effects caused some unwanted visual artefacts causing the location of the template partitions to remain visible even after removing the template. Wall effects were anticipated to be more important in regions of higher permeability (e.g. where class 1 or 2 beads were used). In an attempt to minimize spatial variations of bead diameters – and thus wall effects – at the porous medium/wall interface, a thin (< 1 mm) layer of the finest beads (class 9) was uniformly sprinkled across the flow cell prior to removing the template. Unfortunately, when the template was pulled up, the fine beads located on its walls were displaced into the adjacent blocks,

leaving a clear imprint of the template partitions at the surface of the porous medium. This phenomenon explains why the limits between blocks are still clearly defined even after the template was removed. Notice how the “ghost” partitions between blocks appear darker than surrounding (coarser) blocks in plain light (Figure 2-4(c)). This is due to the fact that the finer bead classes generally appear lighter in colour compared to the coarser beads. Fortunately, this side effect was limited to the surface of the porous medium, and thus became much less apparent when the flow cell was viewed using transmitted light (e.g. see Figure 2-4(d)). Moreover, the image normalization processing step, which divides each image containing a solute plume by a background image where only the porous medium and water are present, further diminishes the “blocky” appearance. See Appendix B for details on image processing.

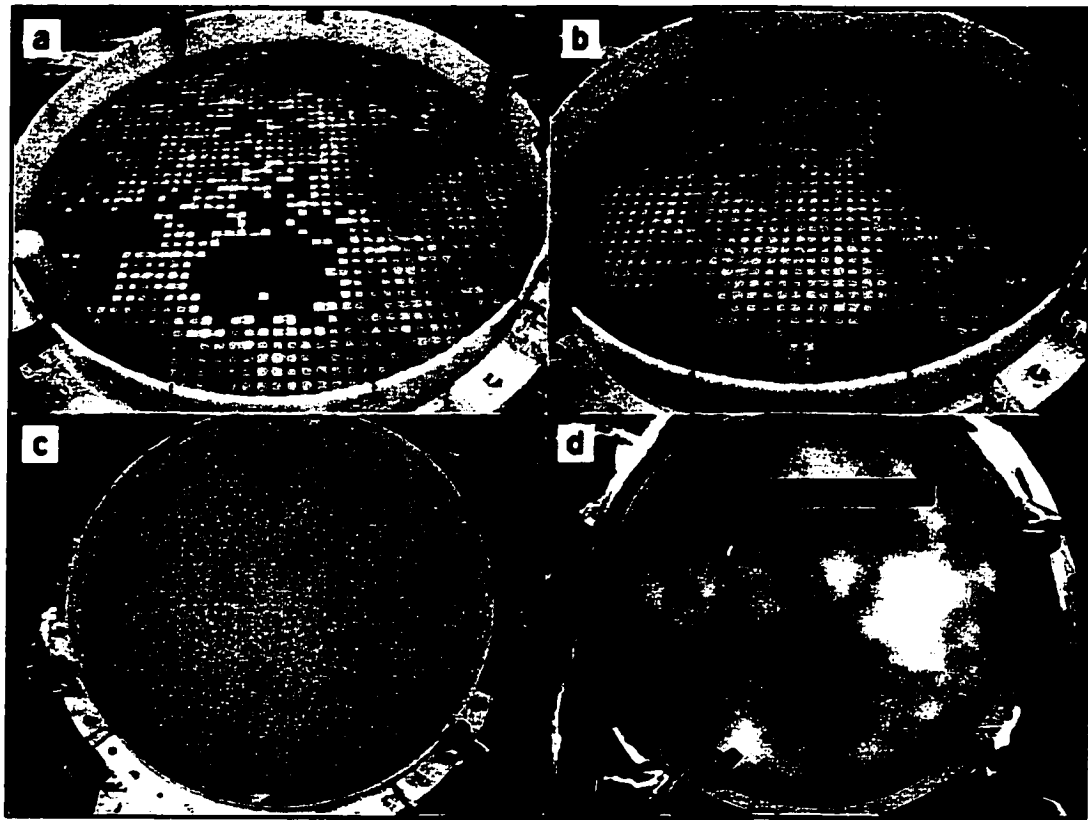


Figure 2-4. Image sequence summarizing the procedure used to construct the heterogeneous porous medium inside the flow cell. Each of the template's $1.5 \times 1.5 \text{ cm}^2$ blocks (about 1450 in total) was filled with a pre-measured volume of glass beads according to the map shown in Figure 2-3. The image in (a) shows the flow cell after bead classes 1-7 had been added. Once all complete blocks (those not partially truncated by the circular outline of the flow cell) were filled (b), the finest beads (class 9) were used to fill the partial blocks around the flow domain. The template was then gently removed (c), and the clear Plexiglas® cover firmly inserted to seal the water-saturated flow cell. The entire assembly was placed upright in front of the light box (d). The entire procedure took place under water to avoid air entrapment within the porous medium.

The heterogeneous nature of the porous medium used in the laboratory experiments presented in this chapter introduced some new limitations which deserve particular attention. These are the result of the large contrast caused by the spatially non-uniform porous medium, combined with the limitations of the image acquisition system, which conspired to compromise image quality to a certain extent.

By increasing the camera lens aperture and/or exposure time, it was possible to “see” through the darker low permeability areas dominated by class 8 or 9 beads, at the expense of over-exposing the most conductive (brighter) regions. If, on the other hand, the camera settings were selected as to fully capture the details in the bright (conductive) zones, contrast was lost in the darker areas. Consequently, preference had to be given to either the zones of highest or lowest permeability (brightest and darkest areas, respectively). Following some experimentation, it was decided to focus on the permeable areas, most likely to host the tracer. Because of the spatial configuration of the permeability field, the finer beads (dark blue shades in Figure 2-2) were generally confined to areas of the flow domain located away from the main solute transport path followed by the tracer. It was therefore most important to visualize the tracer distribution as accurately as possible as it passed through the zones of higher flow rates (red areas in Figure 2-2). However, this bias toward the high permeability areas also introduced additional noise in the low permeability areas, due to their low intensity.

These errors were translated as artificial concentrations, indistinguishable from actual solute concentrations, but were in large part successfully excluded from the moment calculations. This was possible via the image processing plugin, which only incorporated into the spatial moments the contribution from pixel values located within a prescribed local region of interest (ROI), thereby ignoring the artificial (noise-induced) values. Appendix C provides the details on the *Moment_Calculator* software plugin, developed as part of this study.

These contrast-related problems could be avoided in future experiments, in one of two ways. The first would be to choose a spectrum of glass bead sizes that has a smaller range of diameters, to reduce the brightness contrast. For example, this would have been easily achieved by eliminating either the brightest or darkest beads (e.g. classes 1-2 or 8-9, respectively). The other alternative would be to use a digital camera equipped with a solid state sensor (CCD) capable of higher pixel depth. The 8-bit camera used for this study was only capable of producing 256 levels of grey; the expanded contrast of a 10- or 12-bit device would most likely be sufficient to accommodate the contrast range from all 9 bead classes, and possibly more.

Numerical model

The permeability distribution shown in Figure 2-3 was imported into the hydrogeological flow model (Visual MODFLOW). Each row and each column was subdivided into four, so that each of the original blocks (43×43 in total) was split into 16 smaller grid blocks to produce a grid of $172 \times 172 = 29,584$ cells, each $(1.51 \text{ cm} / 4)^2 = 0.143 \text{ cm}^2$. Cells outside the circular flow domain, whose boundaries are outlined using a yellow circle in Figure 2-3, were marked as “inactive” to effectively remove them from the flow and transport simulations. Fixed head (Type I) boundary conditions were assigned to grid cells whose coordinates matched those of the 12 flow cell’s inlet/outlet ports.

Initial concentrations (C_i) were assigned using the map presented in the previous section (Figure 2-1(b)), after re-scaling the initial concentration values to reflect the injection concentration $C_0 = 15.47 \text{ mg L}^{-1}$ in the current set of experiments. Recall that the initial concentration map was estimated by vertically mirroring the visible portion of the early plume to “remove” the injection port covering its left side. Because the area occupied by the plume immediately following injection is small (a diameter of about 3 cm), the plume is limited to a relatively homogeneous portion of the porous medium. Therefore, using the C_i map “engineered” from an experiment that took place in a homogeneous porous medium was assumed to be inconsequential.

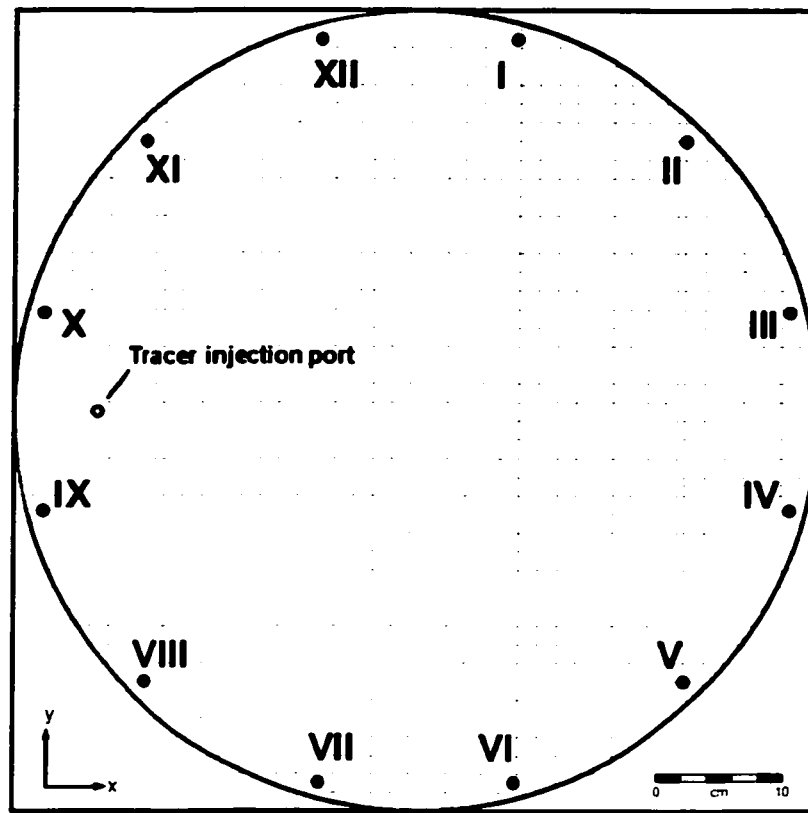


Figure 2-5. Simplified representation of the laboratory flow cell illustrating the configuration of the inlet/outlet ports, shown as black circles and labelled from I through XII. The internal diameter of the flow domain is 65 cm. The boundaries of the template used for the construction of the heterogeneous porous medium are shown by the dotted lines. Each 2.28 cm^2 block within the circular flow cell (approximately 1500 in total) was filled with one of nine size classes of glass beads, as defined from the map shown in Figure 2-3 and whose physical properties are summarized in Table 2-3. Note that for the numerical simulations, the 172×172 cell finite difference grid was created by subdividing each block into 16 cells, each 0.143 cm^2 .

1.3 Description of the flow scenarios

Constant head boundary conditions were specified at each of the 12 ports inside the flow cell. For the steady state cases, the hydraulic heads imposed at each port remained constant throughout the duration of the run, while they were varied in time for the transient scenarios. Flow magnitude and direction were controlled using the hydraulic control system (HCS) described in Appendix A.

Three sets of experiments were conducted, based on the magnitude of the hydraulic gradient: the homogeneous porous medium was subjected to two sets (“low gradient” and “high gradient”), and one set for the heterogeneous medium. For the first set (cases 1-4), the gradient magnitude was maintained constant, with $i = 0.12$, while a value of $i = 0.22$ was used in the second set (cases 5-8); for the third set, the hydraulic gradient was $i = 0.21$ allowing a direct comparison with the second set. The hydraulic gradient was fixed by imposing a constant hydraulic head at each inlet/outlet port location; values are shown in Table 2-4. Note that the hydraulic head values were initially specified at the inlet/outlet ports as shown in Figure 2-6, for all scenarios within a given group of constant hydraulic gradient. The distribution of the hydraulic heads remained unchanged throughout the duration of the steady state flow scenarios (cases 1 and 4). In transient scenarios, on the other hand, the position of the hydraulic heads was changed in time as required by each scenario. The time intervals used for each scenario are shown in Table 2-5. The magnitude of the hydraulic head in any given channel remained unchanged; in order to alter the direction of flow, the hydraulic switch was rotated in increments of 30° to modify the coupling between hydraulic channels and corresponding inlet/outlet ports. For example, consider case 4 in Table 2-1 (reversing flow scenario, with $i = 0.12$) where hydraulic heads were periodically reversed. The direction of the hydraulic gradient was rotated by 180° periodically, resulting in periods of forward and reverse flow. Groundwater flow initially took place towards the right (forward flow). At the end of the first period ($\Delta t_1 = 10$ min), flow was instantaneously reversed so that the hydraulic head distribution became the mirror image (flipped vertically) of what it was during the initial period. This flow reversal marked the beginning of the second period, of length $\Delta t_2 = 5$ min, with the highest head from Table 2-4 (H_1) now assigned to ports III and IV ($h_{III} = h_{IV} = 18.5$ cm), and the lowest head (H_2) to ports IX and X ($h_{IX} = h_X = 11.3$ cm). A similar approach was used for the shifting (zigzag) scenarios, but by rotating the channel/port couplings by 30° or 60° rather than 180° .

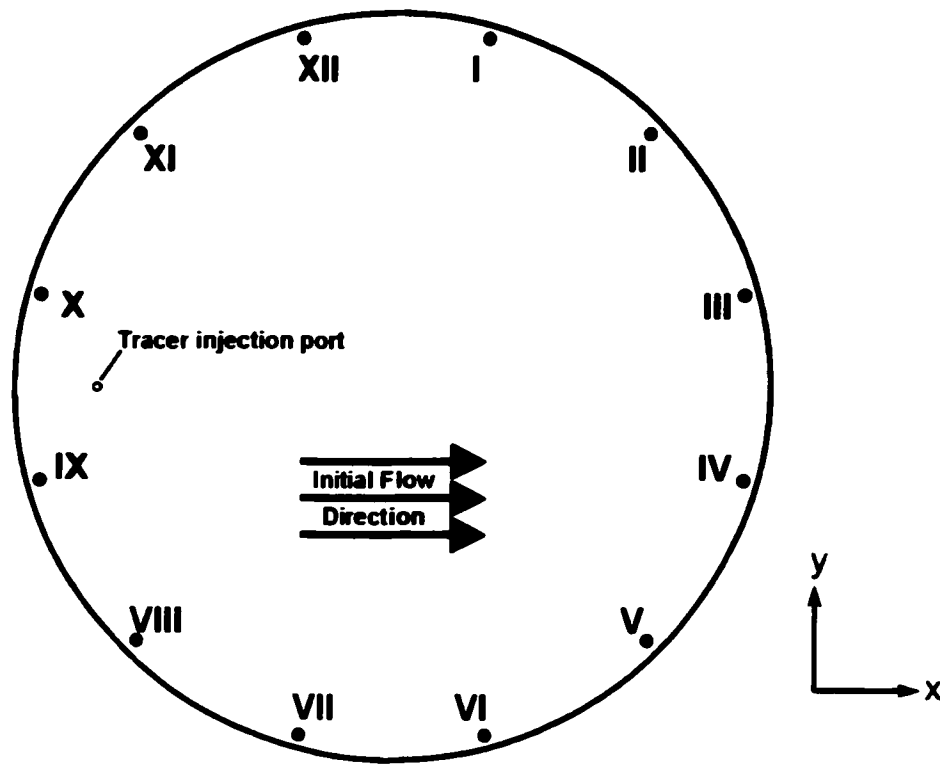


Figure 2-6. Simplified representation of the laboratory flow cell illustrating the configuration of the inlet/outlet ports, shown as black circles and labelled from I through XII. The internal diameter of the flow domain is 65 cm.

Table 2-4. Constant head values imposed at inlet/outlet ports. These were measured in the laboratory using manometers, and were used as input values in the numerical simulations to define the constant head cells. Refer to Figure 2-6 to view the geometric arrangement of the 12 inlet/outlet ports. Cases 1-8 were conducted using the homogeneous porous medium, while cases 9-12 refer to the heterogeneous experiments.

a Level	Position parallel to flow, x (cm)	Hydraulic head, h (cm)			b Inlet/outlet port label
		Homogeneous		Hetero.	
		Cases 1-4 $i = 0.12$	Cases 5-8 $i = 0.22$	Cases 9-12 $i = 0.21$	
H_1	2.36	18.5	22.6	21.7	IX, X
H_2	10.44	17.6	20.0	19.9	VIII, XI
H_3	24.42	15.8	17.0	16.8	VII, XII
H_4	40.58	13.9	13.7	13.5	I, VI
H_5	54.56	12.1	10.8	10.4	II, V
H_6	62.64	11.3	9.15	8.90	III, IV

a. These were maintained for the full duration of the experiments.

b. The location of the inlet/outlet ports is shown in Figure 2-6. The inlet/outlet ports correspond to the hydraulic head levels for the entire duration of the steady state flow scenarios (cases 1, 5 and 9); for transient scenarios (cases 2-4, 6-8 and 10-12), the hydraulic head levels only apply to these specific ports during the initial and final flow periods – for other sub-intervals, the pairs of ports where the hydraulic heads were specified were governed by the requirements of each transient flow scenario (see Table 2-5).

Table 2-5. Schedule used to assign specified hydraulic heads to pairs of inlet/outlet ports. Refer to Figure 2-6 to view the geometric arrangement of the ports, and to Table 2-4 for the hydraulic head imposed at each port ($H_1, H_2, \dots, H_6$). Cases 1-8 were conducted using the homogeneous porous medium, while cases 9-12 refer to the heterogeneous experiments.

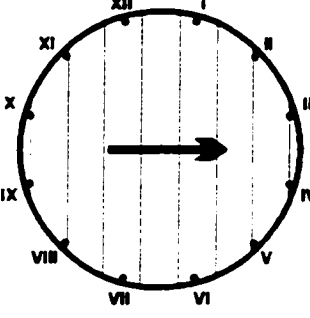
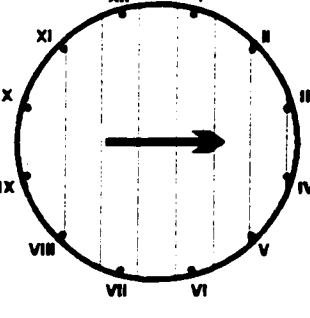
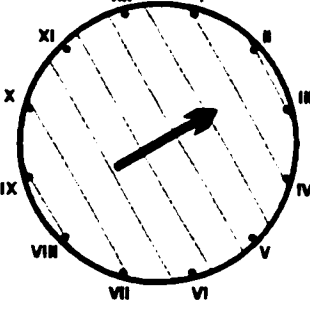
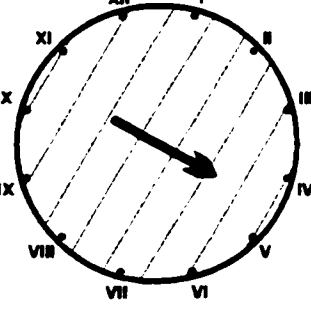
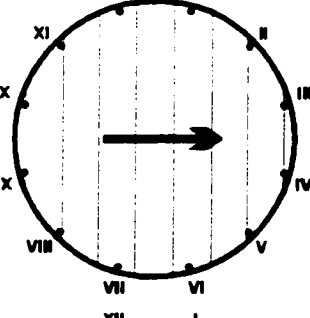
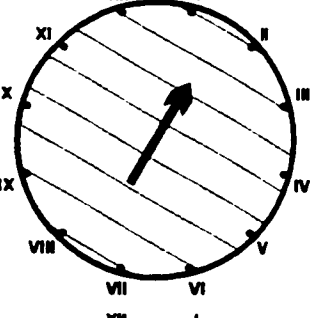
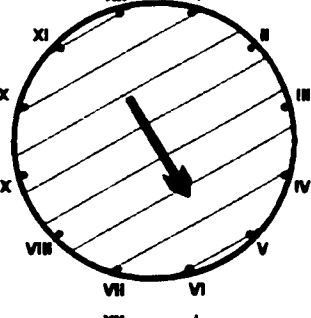
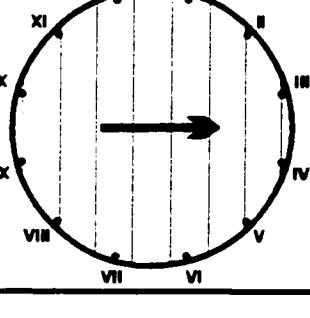
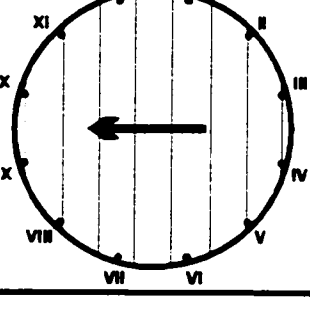
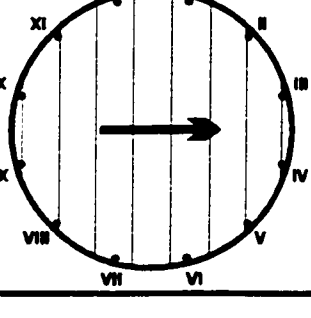
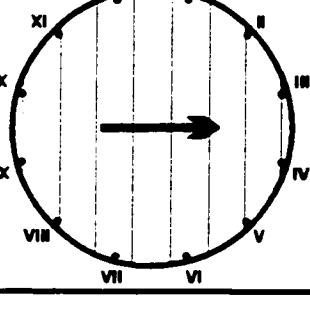
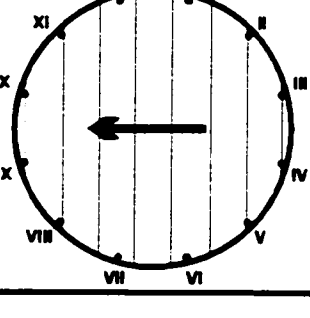
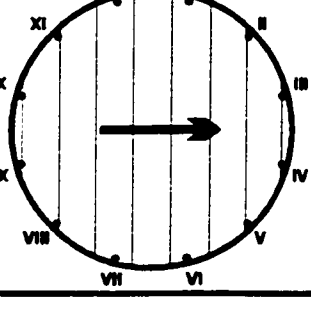
Flow pattern		Period Duration (min)			Inlet/outlet ports where constant heads H_i are applied					
		$i=0.12$	$i=0.22$	$i=0.21$	H_1	H_2	H_3	H_4	H_5	H_6
Steady State	Δt_1	case 1 40	case 5 22	case 9 22	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
Zigzag 30°		case 2 2.5 to 3	case 6 1.5	case 10 2.5	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
	Δt_1	5	5	5	VIII, IX	VII, X	VI, XI	V, XII	I, IV	II, III
	$\Delta t_2 \text{ \& } \Delta t_{f-1}$	10	10	10	X, XI	IX, XII	I, VIII	II, VII	III, VI	IV, V
	$\Delta t_3 - \Delta t_{f-2}$	variable	variable	variable	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
Zigzag 60°		case 3 2 to 3	case 7 1.5	case 11 2.5	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
	Δt_1	2.5	2.5	2.5	VII, VIII	VI, IX	V, X	IV, XI	III, XII	I, II
	$\Delta t_2 \text{ \& } \Delta t_{f-1}$	5	5	5	XI, XII	I, X	II, IX	III, VIII	IV, VII	V, VI
	$\Delta t_3 - \Delta t_{f-2}$	variable	variable	variable	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
Reversing		case 4 10	case 8 5	case 12 5	IX, X	VIII, XI	VII, XII	I, VI	II, V	III, IV
	$\Delta t_1, \Delta t_3, \dots, \Delta t_f$	5	2.5	2.5	III, IV	II, V	I, VI	VII, XII	VIII, XI	IX, X

Table 2-4 shows the six constant head values that were used for each series of experiments: each pair of ports was assigned a specific head as required by the desired flow direction at any given time. For instance, in order to generate a uniform flow field of desired magnitude in the positive x direction (see Figure 2-6), the highest constant head value from Table 2-4 was assigned to the ports labelled **IX** and **X**, while the lowest head value was imposed to ports **III** and **IV**. For the first set (cases 1 to 4), the gradient magnitude was 0.12, with constant heads set to values ranging between 0.185 and 0.113 m; for the second series (cases 5 to 8), the mean gradient was 0.22.

Flow direction was modified periodically during the course of the experiments, with the exception of the steady state scenarios (cases 1 and 5), where the mean flow direction remained constant in the positive horizontal (x) direction. For the transient cases, flow direction was rotated relative to the longitudinal direction (positive x -axis); these include cases 2 and 3 ($\pm 30^\circ$ changes in flow direction), and cases 6 and 7 ($\pm 60^\circ$ variations). For the reversing flow cases (4 and 8), the hydraulic gradient was alternated between the

positive and negative x directions. Each case was defined by the combination of the two principal factors under investigation, namely gradient magnitude and direction. Because changes in bulk flow conditions occurred almost instantaneously, the transient experiments could be viewed as a succession of steady flow conditions, with mean gradient magnitude and direction both remaining unchanged within each sub-interval.

Table 2-6. Schematic representation of the flow configurations from all flow scenarios. The hydraulic heads imposed at each inlet/outlet port (shown as 12 small circles and labelled I through XII below) are shown in Table 2-4, while the schedules used for each flow pattern are shown in Table 2-5. Each period was identified as Δt_f , where f represents the flow period index, from initial ($f = 1$) to final ($f = F$).

Scenario	Time period		
	$\Delta t_1, \Delta t_F$	$\Delta t_2, \Delta t_4, \dots, \Delta t_{F-2}$	$\Delta t_3, \Delta t_5, \dots, \Delta t_{F-1}$
Steady State (cases 1, 5, 9)			
Zigzag 30° (cases 2, 6, 10)			
			
			
Reversing (cases 4, 8, 12)			

1.4 Darcy's Law: validity range

In contrast to field situations where groundwater flow conditions are beyond the experimenter's control, laboratory experiments or numerical simulations allow one the luxury of controlling many of the parameters that regulate both flow and transport. For example, selection of the fluid, granular materials or boundary conditions greatly influences the conditions under which the system allows groundwater to flow and carry solutes. One of the most important considerations that need to be addressed is whether Darcy's Law is applicable under a particular set of conditions.

As discussed above, Darcy's Law is the equation that governs groundwater flow in porous media. It is a linear law that is only valid when flow is laminar and pore water velocity (specific discharge) is neither below nor above threshold values [*Freeze and Cherry, 1979*]. Below a certain threshold, the hydraulic gradient is so weak that flow will not take place.

At the other end of the spectrum is the situation where groundwater flow occurs at such a high rate that it becomes turbulent, and Darcy's Law is no longer applicable. The Reynolds number (Re) is a dimensionless number that expresses the ratio of inertial to viscous forces. It can be used to distinguish between laminar flow occurring at low velocities (where Darcy's Law is valid), and turbulent flow. *Bear [1972]* states that "*Darcy's Law is valid as long as the Reynolds number based on average grain diameter does not exceed some value between 1 and 10*" (p. 126).

The Reynolds number was estimated to ensure that the velocities imposed on the system during the course of the experiments conducted as part of this work were within the validity range of Darcy's Law. Using the diameter from the coarsest granular materials used in this study ($d = 0.05$ cm) and the highest hydraulic gradient ($i = 0.22$) provided the highest expected velocities. Results showed that the velocities in this study are well below the threshold values defined as the upper acceptable limits of validity of Darcy's Law. This was confirmed by Sichardt's empirical formula for borderline gradient. Appendix E provides the details of these calculations.

2 IMAGE PROCESSING

The temporal evolution of tracer clouds migrating through the flow cell under various groundwater flow scenarios was characterized using digital images. The images provided instantaneous snapshots of the spatial distribution of the dissolved tracer plumes. Because images were used as evidence in the subsequent quantitative analyses, a rigorous procedure was established to improve their quality and determine the solute concentration distribution. This process is referred to as image processing (IP). Accurate representation of the solute plumes everywhere in the flow domain then allowed direct comparison of the hydrodynamic dispersion induced by the flow regime in the various experiments. The IP procedure is summarized in Figure 2-7. Refer to Appendix B for a detailed description of the steps involved. The purpose of the procedure is to produce a distribution of solute concentration values from the grey levels measured by a monochrome digital camera. It is an adaptation of the technique proposed by *Schincariol et al.* [1993].

Image Processing Procedure

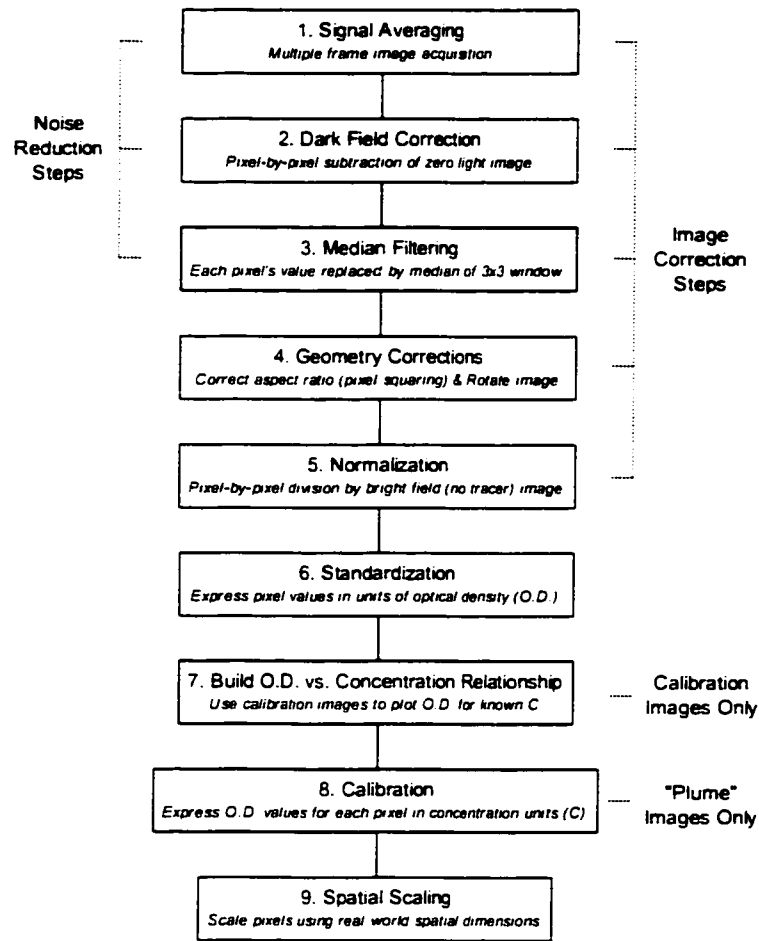


Figure 2-7. Overview of the image processing steps. Steps 1 through 7 were applied to all images, while the procedures in steps 8 and 9 were necessary only for calibration or plume images, respectively. See text for details.

Through a series of image processing operations, the grey levels contained in the photographs were converted into solute concentrations. Raw images were first corrected to remove defects such as noise and other imperfections. This was achieved through the first three steps in Figure 2-7. In order to account for the rectangular aspect ratio of the elements on the camera's sensor, the image was resized in an operation called "pixel squaring". Images were also rotated as required to align the reference points marked on the front cover of the flow cell. The images were then normalized to eliminate undesirable brightness variations present in the background. These imperfections were

due to a combination of factors that may include, for example, non-uniform illumination, packing density variations or impurities. Images that contained the tracer were normalized via a pixel-by-pixel division by the corresponding “background” image, containing only glass beads and water. This operation is illustrated in Figure 2-8 for typical images taken from the laboratory experiments discussed in the following chapters. The standardized images shown on the left were obtained from one homogeneous (a) and one heterogeneous scenario (d). They were expressed relative to the background images shown in the central column ((b) and (e)) to produce the normalized images on the right ((c) and (f)). Notice that the spatial variations in brightness are almost completely eliminated by this operation. In fact, the tracer plume is virtually indistinguishable from the surrounding medium in Figure 2-8 (d) but clearly stands out following normalization (Figure 2-8 (f)).

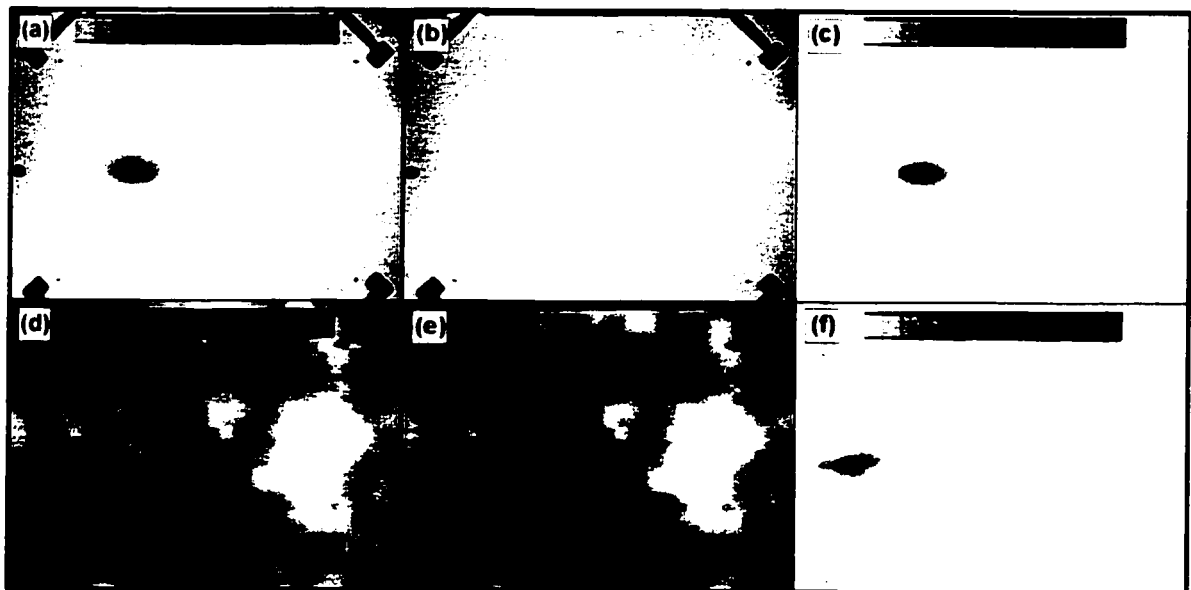


Figure 2-8. Effect of normalization, illustrated for a homogeneous (top) and a heterogeneous (bottom) porous medium. A standardized image containing a plume migrating through a homogeneous porous medium (a) is divided pixel by pixel by the bright field image (b). In the resulting normalized image (c), the zero-concentration (background) is more uniform. In the case of a heterogeneous porous medium (d), it is very difficult to distinguish the solute plume from the background. After division by the bright field image (e), the resulting normalized image (f) shows the tracer distribution very clearly. Each image is 52.8 cm wide by 39.5 cm high.

The normalized images were then standardized to minimize variations in overall lighting conditions between experiments. This was achieved by converting pixel values into units of optical density (OD) using a standard stepped tablet present at the top of all images, away from the plume migration path. The standardization process applied a curve fitting function to relate the normalized grey values at each pixel to standard units measured from the stepped wedge. The standardization operation was also necessary as a way to express pixels on a linear scale, allowing the pixel values to be related to actual tracer concentrations through calibration. To convert pixel values, now in units of OD, in units of tracer concentration (in mg L^{-1}), the OD distribution from a laboratory experiment (homogeneous steady state, case 1) was calibrated using the concentrations predicted from the corresponding numerical simulation were analyzed by computing the spatial moments up to fourth order.

3 SPATIAL MOMENTS

Spatial moments up to second-order have been used extensively to investigate the advective and dispersive behaviour of solute plumes for several large-scale field experiments. Notable examples include the CFB Borden site, Ontario [e.g. *Freyberg*, 1986; *Rajaram and Gelhar*, 1991; *Farrell et al.*, 1994a; *Rivett et al.*, 1994a], the Columbus Air Force Base site, Mississippi [e.g. *Adams and Gelhar*, 1992], and the site located near Cape Cod, Massachusetts [e.g. *Garabedian et al.*, 1991; *Kapoor and Gelhar*, 1994]. Moments are also commonly used to depict the concentration distribution resulting from numerical simulations of contaminant transport in groundwater systems [e.g. *Rubin and Bellin*, 1994; *Sposito and Dagan*, 1994; *Zhang and Chi*, 1995; *Indelman and Rubin*, 1996; *Srivastava and Brusseau*, 1996; *Naff et al.*, 1998b; *Russo*, 1998].

In this study, spatial moments calculated from the concentration distributions derived from calibrated digital images were used to characterize the tracer plumes at each time step. Their temporal evolution provided a basis for analyzing the significance of flow transience on contaminant spreading in granular media. This technique was used to visualize the effects of flow variations on the movement of the dissolved RWT tracer as it travels inside the flow cell through the porous medium. The concentration distributions

derived from the numerical simulations were also described from the spatial moments. The equations used to derive spatial moments, as well as other useful parameters derived from them, are presented in Appendix C.

The general formula used to calculate the spatial moments from a concentration distribution C in up to three dimensions (x, y, z) is given by [e.g. *Freyberg, 1986*]:

$$M_{ijk}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) x^i y^j z^k dx dy dz \quad (2.2)$$

where n is the porosity of the porous medium and the order of the moment is given by $i+j+k$.

In the context of this work, we are interested in describing concentration distributions in both space and time of a dissolved plume. This was achieved by calculating the zero-order- to fourth-order spatial moments from the solute plumes. For the experimental data set, spatial moments were estimated using a plugin extending the functionality of the IP software package ImageJ was developed for this study (see details in Appendix C). An existing code [*Burr et al., 1994*] was adapted to compute the spatial moments from the numerical dataset. The interpretation of the zero- to fourth-order spatial moments and their derivatives are summarized in Table 2-7.

Table 2-7. Interpretation of spatial moments and their time derivatives.

Order	Interpretation	Time derivative
0	Total mass of dissolved solute	Rate of mass gain/loss
1	Location of centre of mass (centroid)	Velocity of centroid (equivalent to the groundwater velocity in this case)
2	Dispersion (spatial variance) around centroid Orientation (with respect to the longitudinal direction, x) Elongation (aspect ratio of long to short plume axes)	Rate of spreading related to the dispersion coefficient and macrodispersivity
3	Coefficient of skewness (degree of symmetry around centroid, compared to a normal (Gaussian) distribution)	
4	Kurtosis (degree of flatness or peakedness, compared to normal (Gaussian) distribution)	

4 MACRODISPERSIVITY

Macrodispersion, in lay language, can be thought of as the process by which contrasts in solute concentrations are mixed with ambient groundwater. It is described more rigorously by *Bear* [1972] as a “... nonsteady, irreversible process”, whereby “... as flow takes place the tracer gradually spreads and occupies an ever-increasing portion of the flow domain, beyond the region it is expected to occupy according to the average flow alone.” (pp. 579-580). Generally, dispersion, described by a coefficient of dispersion D , is conceptualized as the result of mechanical mixing and molecular diffusion D_m .

Conceptually, macrodispersivity is a property of the porous medium alone. It attempts to describe the spatial variability of the flow field at scales smaller than the representative elementary volume (REV). In practice, however, it is observed that dispersivity depends on the size of the REV, on the magnitude of the average velocity, and on the temporal variability of the flow field – the focus of this research. Macrodispersivity is described briefly below, and in detail in Appendix D.

The dispersion coefficient D_{ij} of the solute plume (a tensor) may be defined from its variance σ_{ij}^2 from *Aris* [1956]:

$$D_{ij} = \frac{1}{2} \frac{d}{dt} \sigma_{ij}^2(t) \quad (2.3)$$

The relationship in between dispersion and the time rate of change of the covariance tensor assumes a Fickian (constant) dispersive process. The hydrodynamic dispersion tensor can also be described by *Bear* [1972]:

$$D_{ij} = D_m \mathbf{I} + A_{ij} \|\mathbf{v}\|, \quad (2.4)$$

where $\mathbf{I}$ is the identity matrix, A_{ij} is the macrodispersivity tensor, and $\|\mathbf{v}\|$ is the magnitude of the velocity vector. At the scales of interest in this study, mechanical mixing is expected to be the dominant process, and the contribution of D_m to dispersion is considered negligible. Because in advection-dominated problems diffusion is often very small compared to the second term in (2.4), the first term can generally be neglected [e.g.

Freyberg, 1986]. Using this assumption, the apparent macrodispersivity tensor can be obtained by combining (2.3) and (2.4):

$$A_{ij} = \frac{1}{\|\mathbf{v}\|} D_{ij} \quad (2.5)$$

The term “apparent macrodispersivity” is used [as in e.g. *Freyberg, 1986; Farrell et al., 1994a*] to distinguish the quantity in (2.5) from a “true macrodispersivity”, which is used to describe the behaviour of solute plumes that approximates a Gaussian (normal) distribution [*Loaiciga, 1988*]. Note that in this study, the word macrodispersivity is used for simplicity, but implies the “apparent” qualifier.

Four methods were identified that could be used to estimate the macrodispersivity, based on the selection of the “appropriate” time interval to evaluate the time derivatives of $\sigma_{ij}^2(t)$ and $\bar{x}_i(t)$. The terms in (2.5) are calculated from time derivatives of quantities derived from spatial moments. That is, A_{ij} is calculated from the slopes of the time-plots of the plume’s centroid location ($\bar{x}_i(t)$) and from its spatial variance ($\sigma_{ij}^2(t)$). Appendix D describes and discusses the four methods: *Global, Stepwise, Absolute Stepwise* and *Finite Difference*. As will be discussed later, the *Global, Stepwise* and *Finite Difference* methods produced nearly identical results, whereas the *Absolute Stepwise* method produced very different results when flow direction variations were present.

CHAPTER 3 – Experimental and Numerical Results for Homogeneous Porous Media

“Experience has shown that science frequently develops most fruitfully once we learn to examine the things that seem the simplest, instead of those that seem the most mysterious.”

– Marvin Minsky

The main objective of this study is to quantify the contribution of time-variations of the flow to solute dispersion in saturated porous media. Since the focus is on the dispersion process, the confounding effects of solute-solid interactions were circumvented by using Rhodamine, a tracer that did not significantly interact with the porous medium (the selection and detailed properties of the tracer are discussed in Appendix A). The thesis examines the dispersive behaviour of two types of porous media; one homogeneous and isotropic, and the other heterogeneous and statistically isotropic. This chapter presents the results for the homogeneous cases while the following chapter presents the heterogeneous cases.

The investigation was conducted using a physical (laboratory) model, and a numerical model. The details of the experimental procedure are given in Chapter 2, including descriptions of the laboratory and numerical models. The reader is referred to Appendix A for a detailed description of the experimental apparatus and to Appendix B for the details of the image processing procedure. The spatial moments computation methods are given in Appendix C and the methods used to compute macrodispersivities are presented in detail in Appendix D.

The four scenarios that were considered can be distinguished by the nature of the flow field:

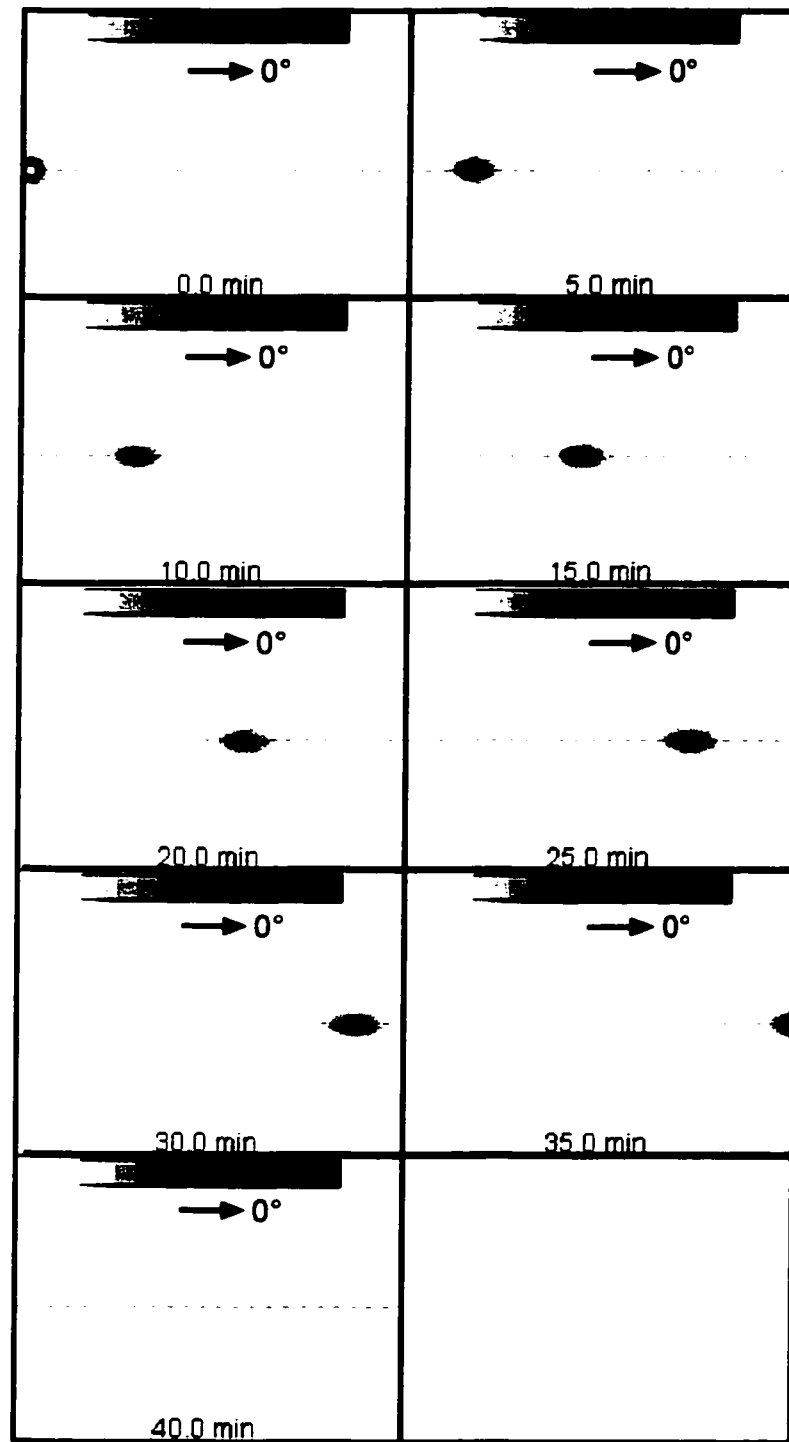
- a) **Steady state unidirectional;**
- b) **Periodic 30° deviations from the average flow direction;**
- c) **Periodic 60° deviations from the average flow direction; and**
- d) **Periodic flow reversals (with net forward flow)**

In addition, two hydraulic gradient magnitudes were considered for each scenario. The effects of the magnitude were examined only for the homogeneous porous medium (this chapter).

1 SPATIAL MOMENTS AND DERIVED QUANTITIES

The details from each experimental run are presented in Appendix G, as time-plots of the parameters derived from spatial moments of orders 1 to 4. The physical interpretation of these quantities was summarized in the preceding chapter.

Each of the eight flow scenario/gradient magnitude experiments was repeated a (varying) number of times. The complete data set is given in Appendix G. Typical runs from the four flow scenarios with the lower hydraulic gradient were selected as illustrative examples for this discussion, and the results are presented over the next several pages. For each of these four sample scenarios, a sequence of images is presented showing frames at five minute intervals followed by a set of graphs presenting the complete time series for the parameters derived from the image sequences. To facilitate comparisons, the experimental results are plotted on the left and the numerical results on the right.



(a)

Figure 3-1. Selected images (a) from a typical steady state scenario (case 1, run 1), accompanied in part (b) to part (g) by the graphs showing the spatial moments derived from this image sequence and from the corresponding numerical simulation. Flow direction is shown by the arrows (angle expressed relative to x). The dashed line is aligned with the injection port and provides a visual reference of the flow "centre line" along x .

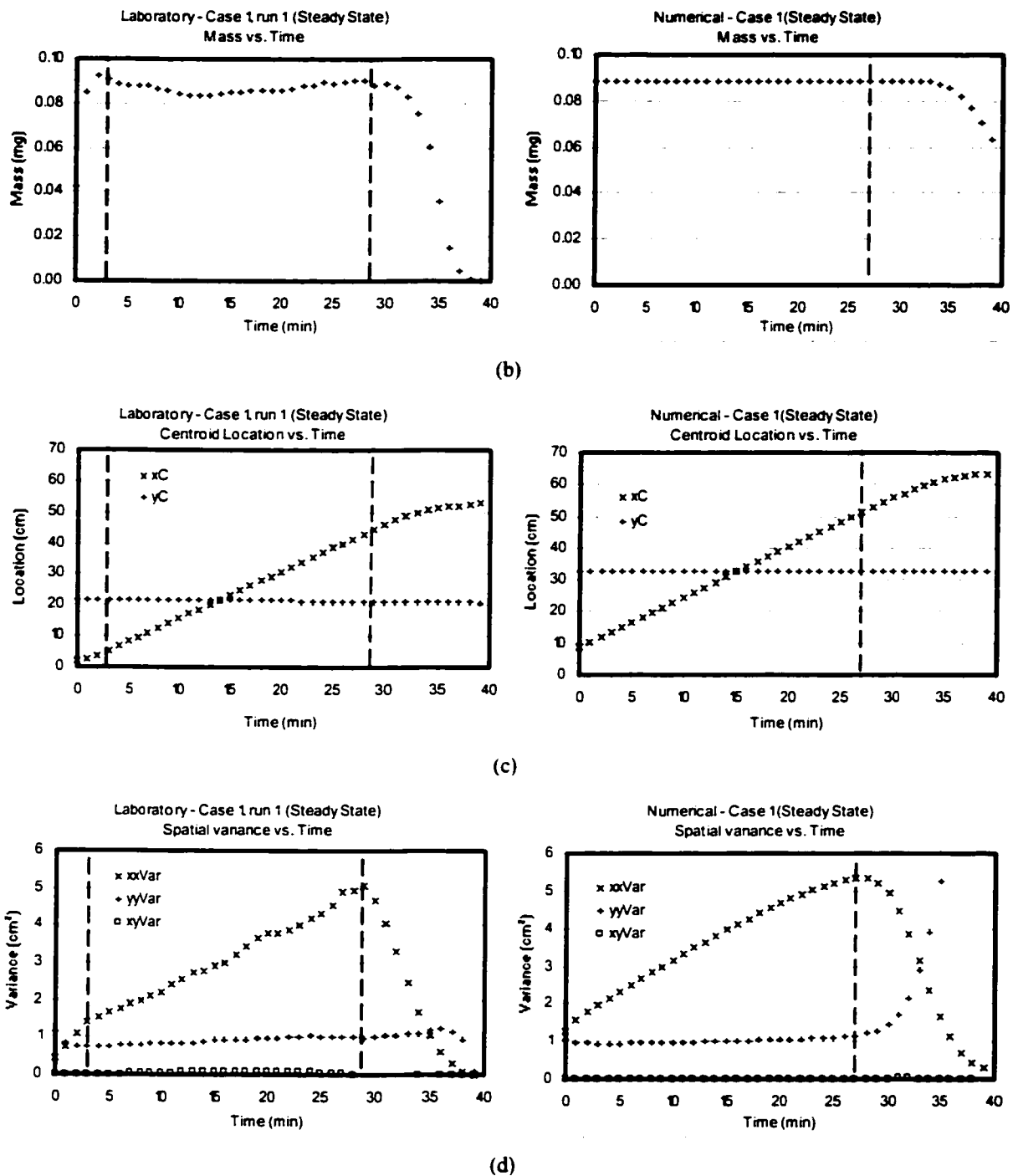
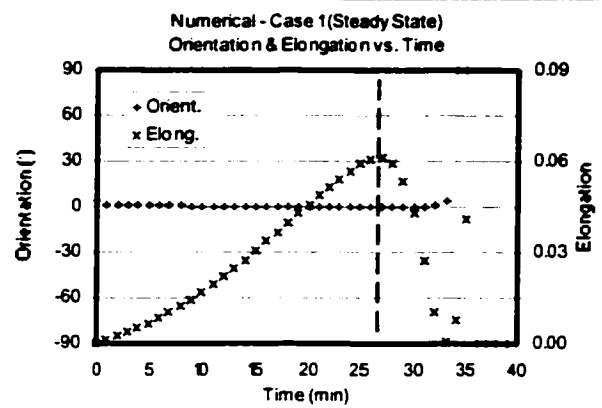
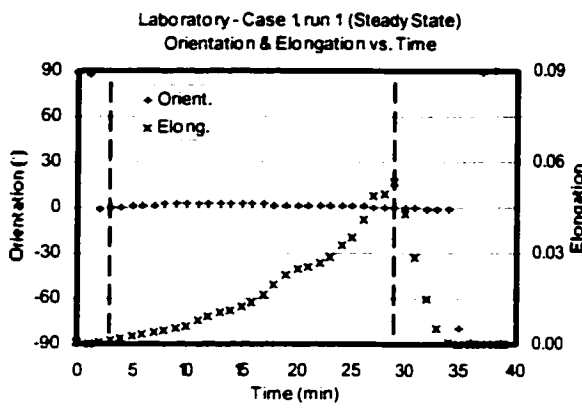
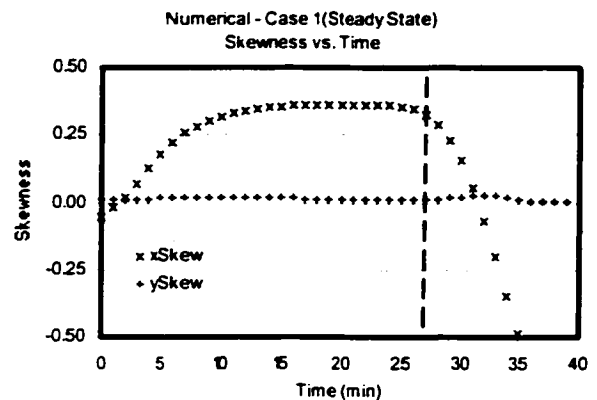
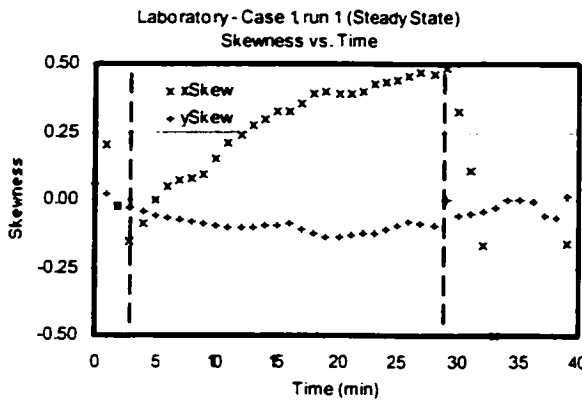


Figure 3-1. (continued)

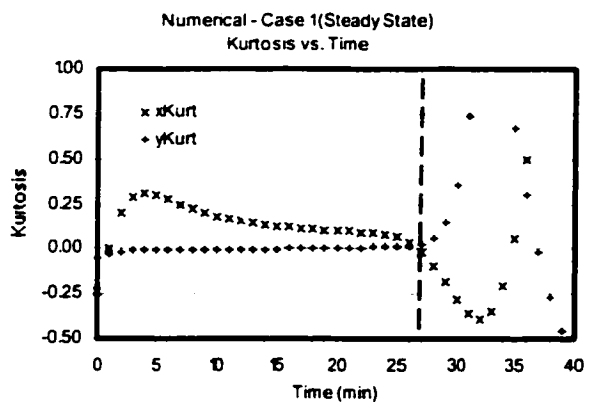
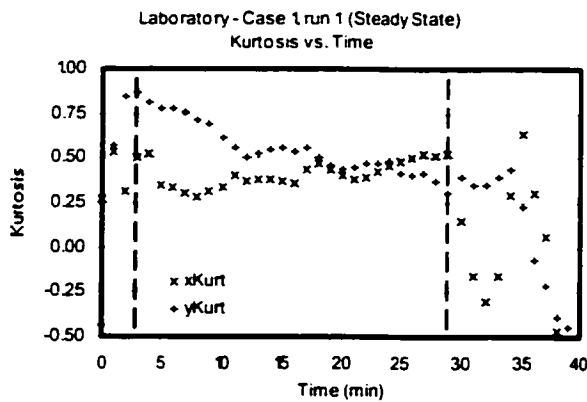
Sample charts showing laboratory (left) and numerical (right) results for case 1 (steady state flow, $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).



(e)



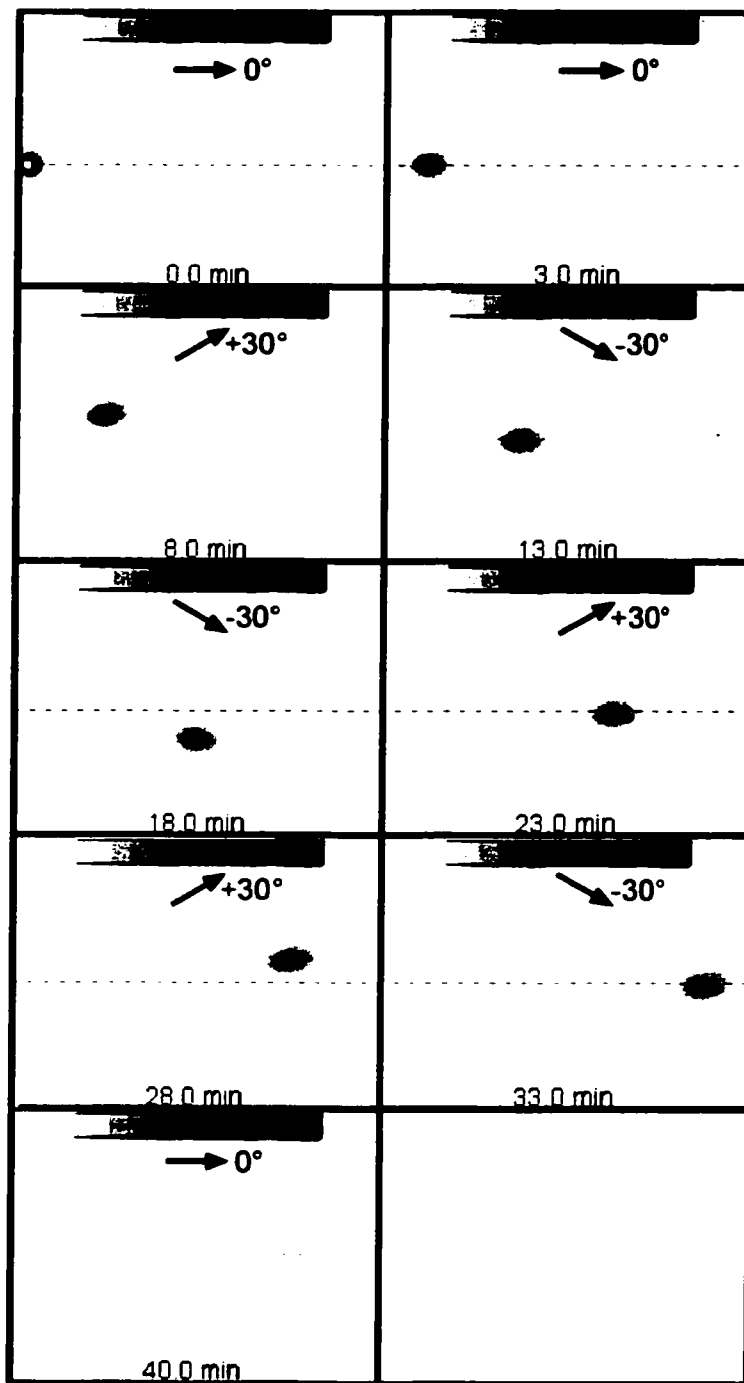
(f)



(g)

Figure 3-1. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 1 (steady state flow, $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).



(a)

Figure 3-2. Selected images (a) from a typical “zigzag 30°” scenario where the hydraulic gradient direction was varied in time at an angle of $\pm 30^\circ$ (case 2, run 3), accompanied in part (b) to part (g) by the graphs showing the spatial moments derived from this image sequence and from the corresponding numerical simulation. Flow direction is shown by the arrows (angle expressed relative to x). The dashed line is aligned with the injection port and provides a visual reference of the flow “centre line” along x .

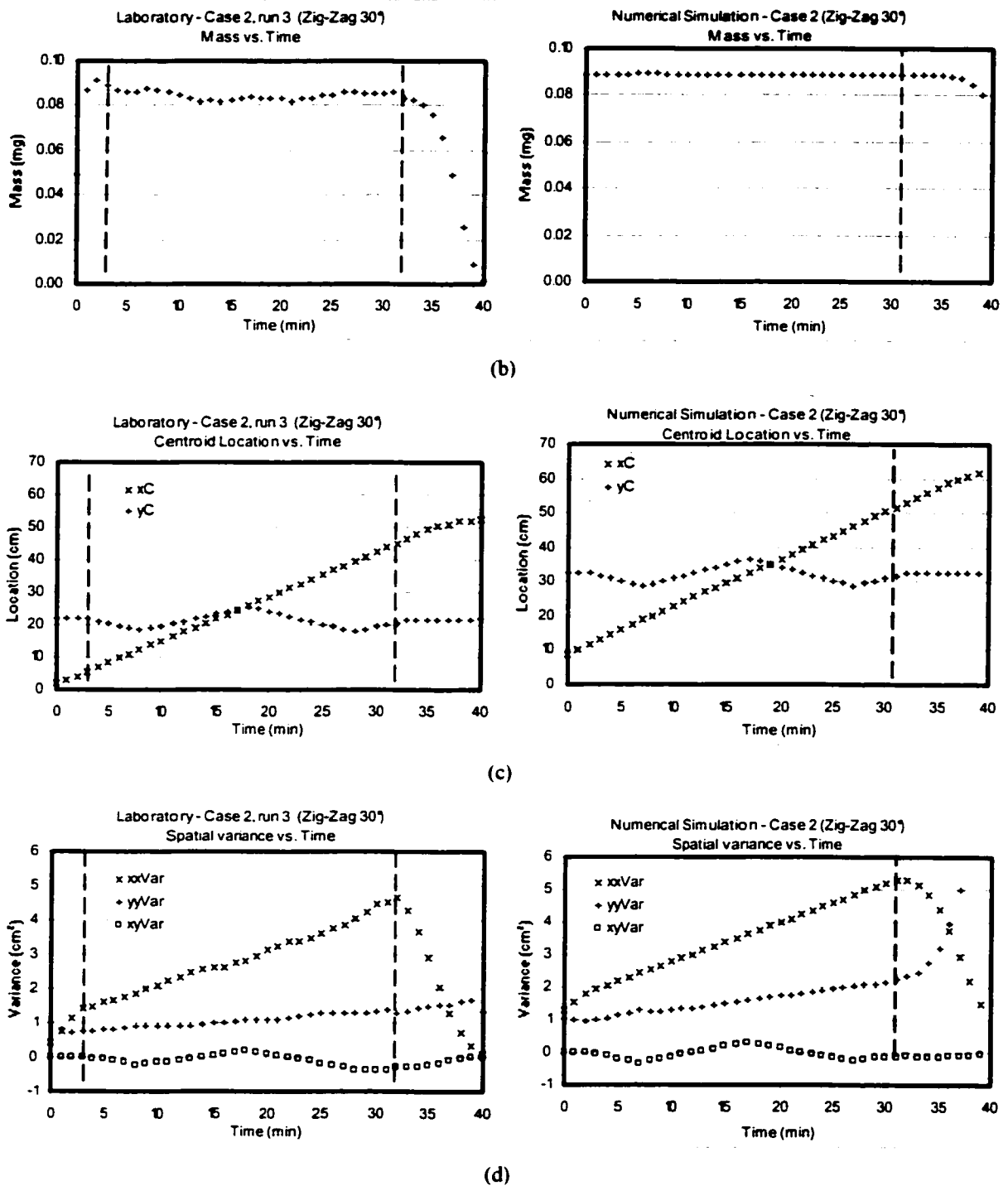


Figure 3-2. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 2 (zigzag flow pattern $\pm 30^\circ$ from longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

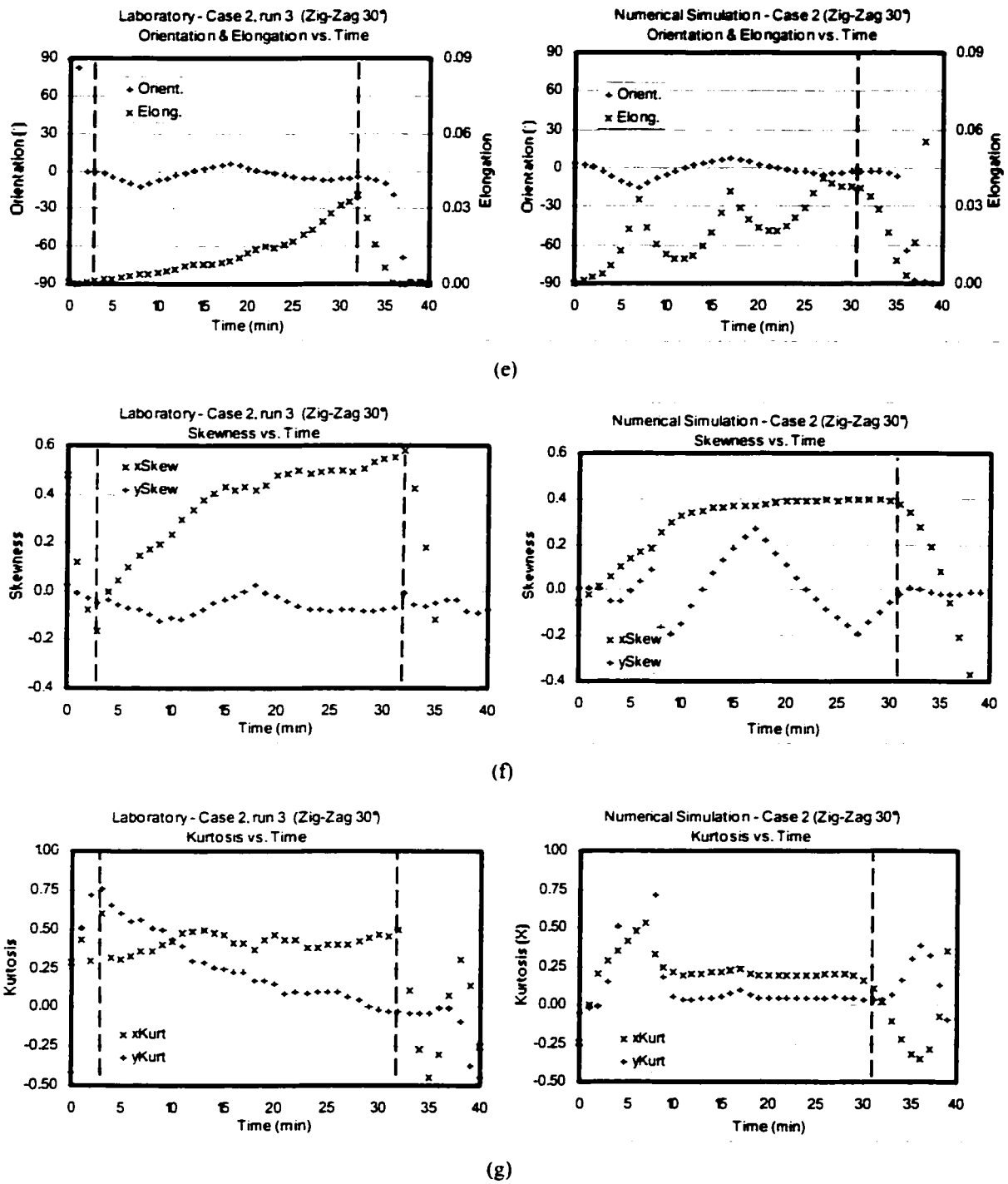


Figure 3-2. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 2 (zigzag flow pattern $\pm 30^\circ$ from longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

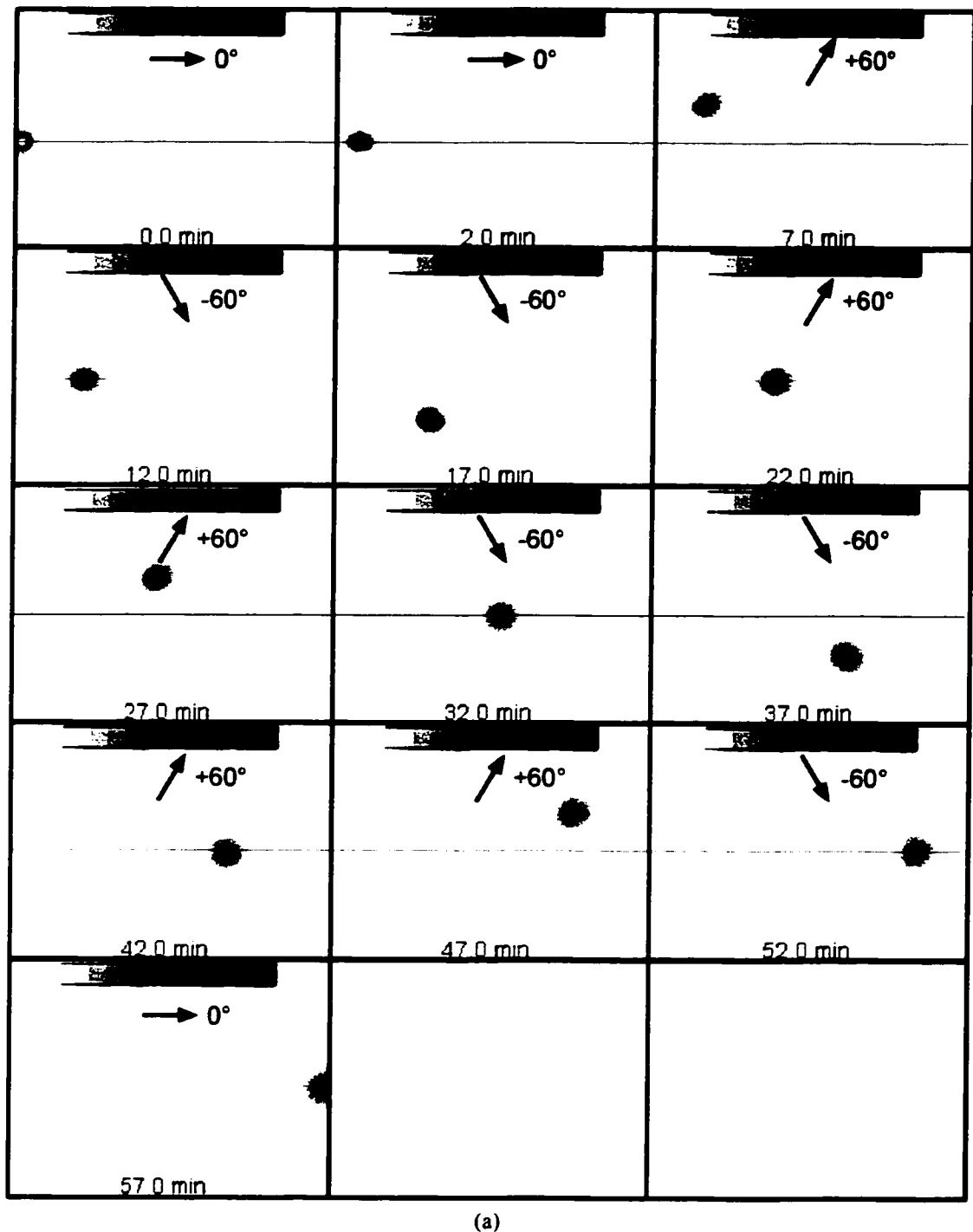
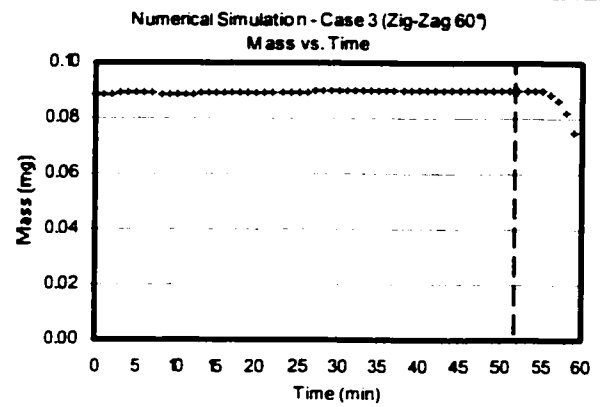
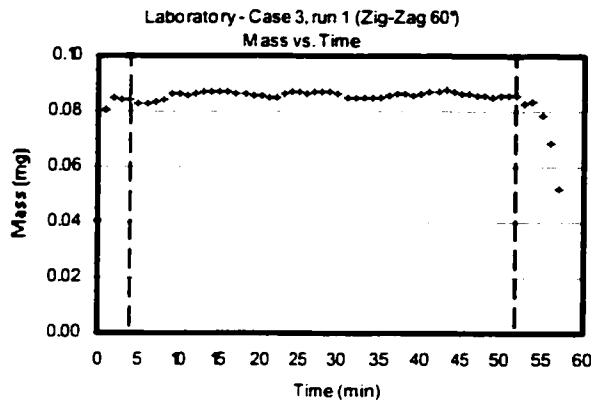
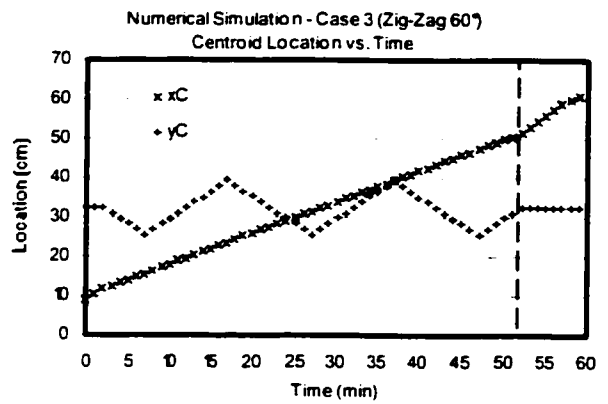
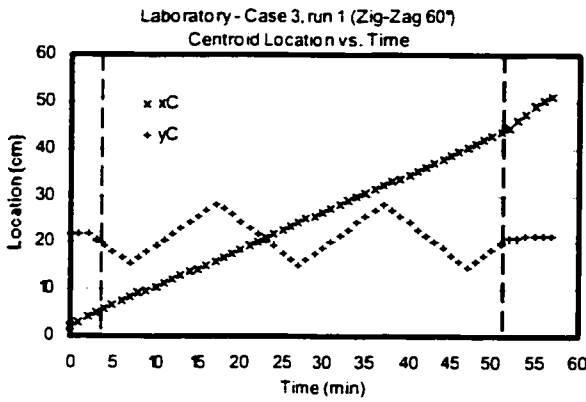


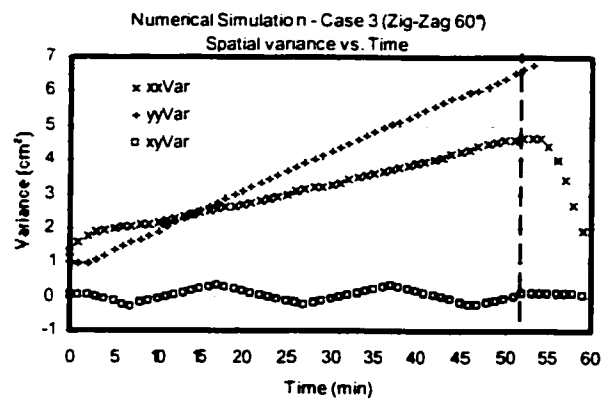
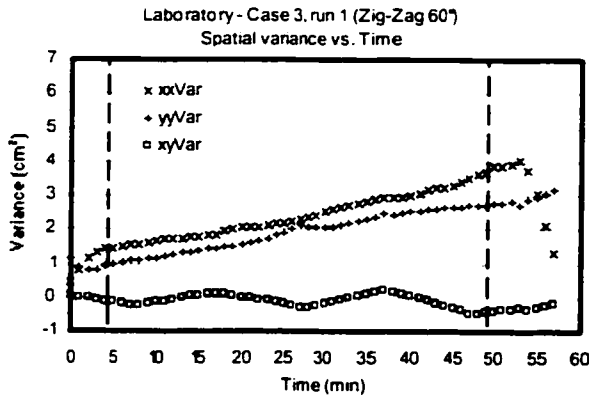
Figure 3-3. Selected images (a) from a typical "zigzag 60°" scenario where the hydraulic gradient direction was varied in time at an angle of $\pm 60^\circ$ (case 3, run 1), accompanied in part (b) to part (g) by the graphs showing the spatial moments derived from this image sequence and from the corresponding numerical simulation. Flow direction is shown by the arrows (angle expressed relative to x). The dashed line is aligned with the injection port and provides a visual reference of the flow "centre line" along x .



(b)



(c)



(d)

Figure 3-3. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 2 (zigzag flow pattern $\pm 30^\circ$ from longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

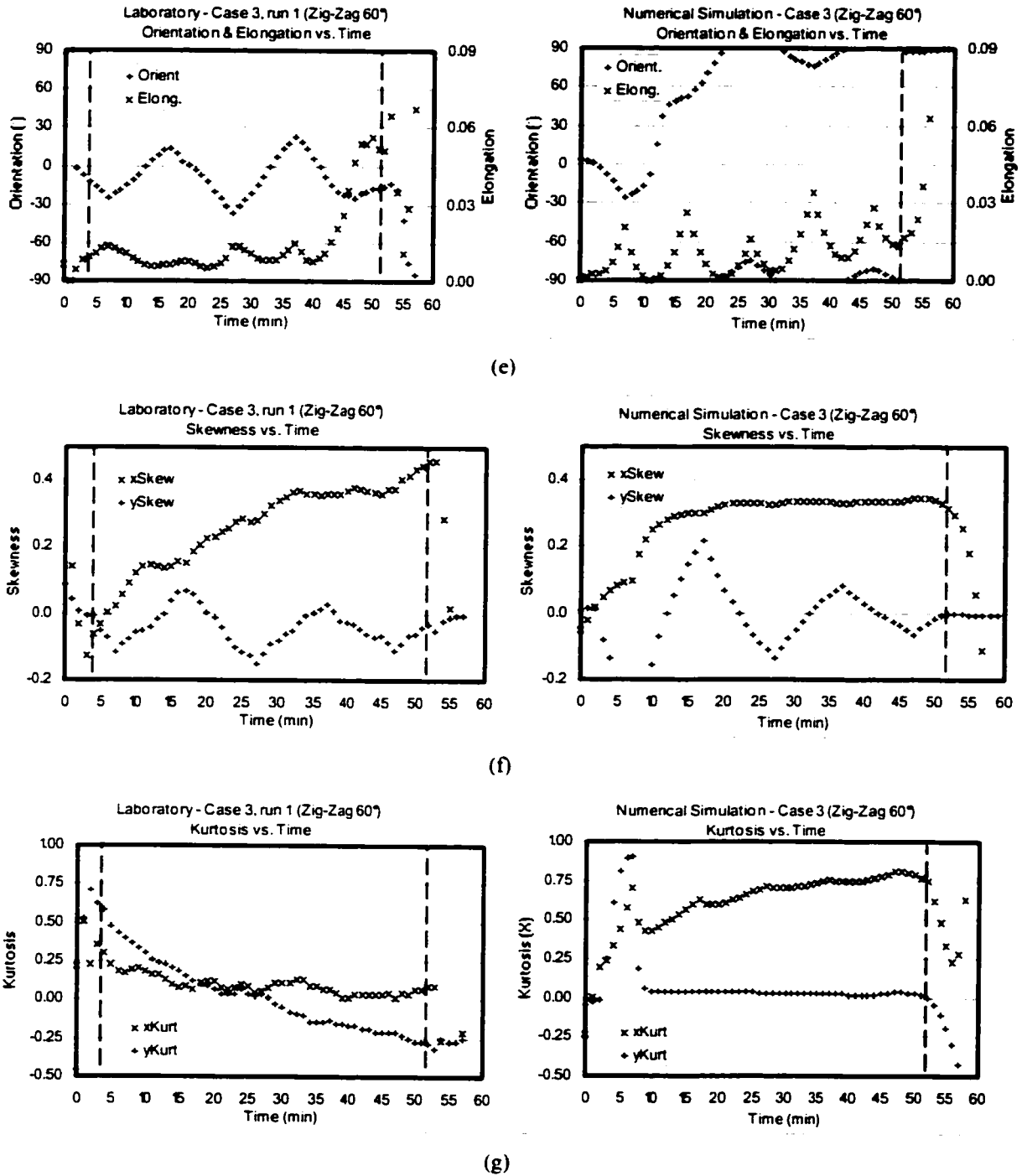


Figure 3-3. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 2 (zigzag flow pattern $\pm 30^\circ$ from longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

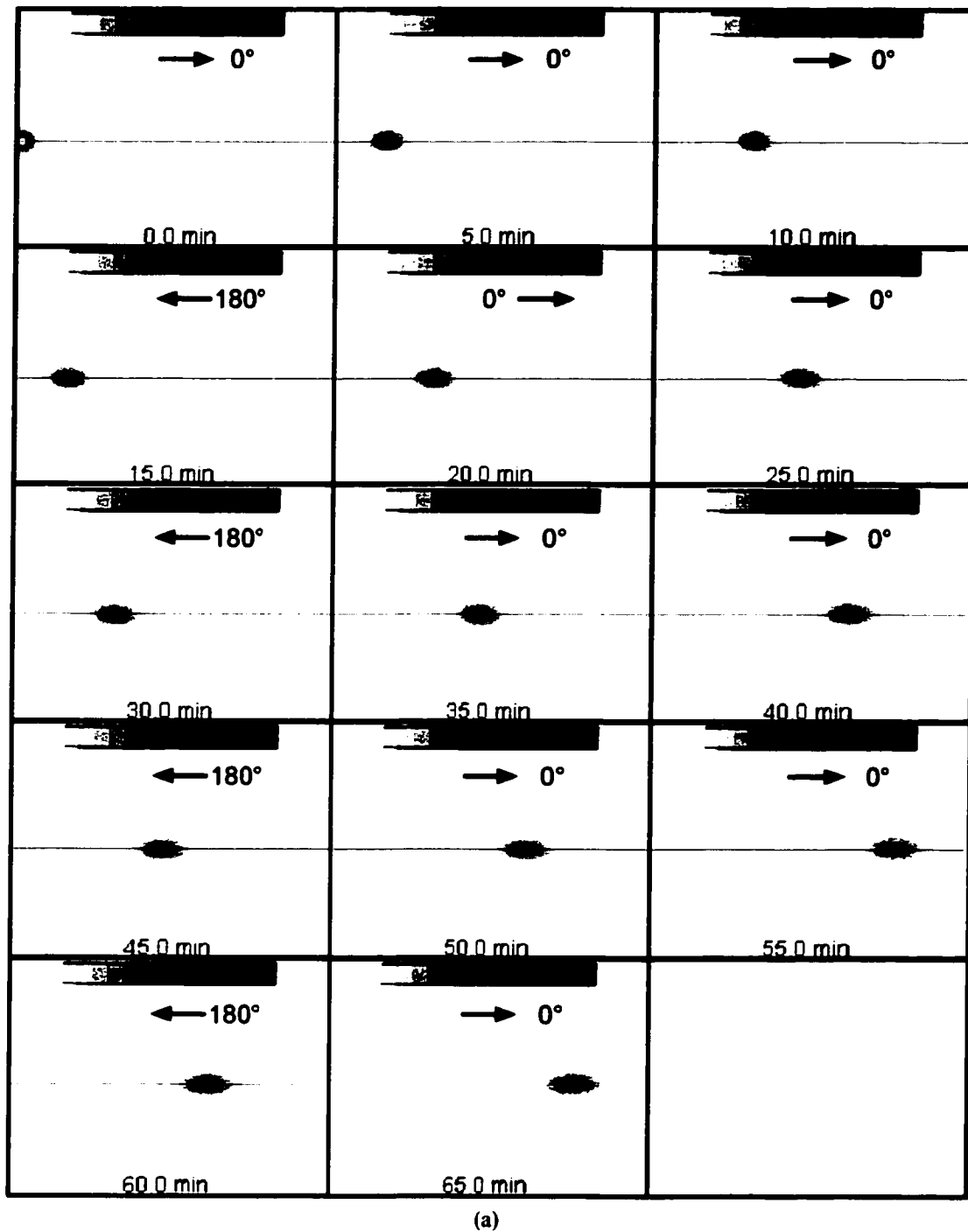


Figure 3-4. Selected images (a) from a typical “reversing” scenario where the hydraulic gradient direction was reversed periodically (case 4, run 1), accompanied in part (b) to part (g) by the graphs showing the spatial moments derived from this image sequence and from the corresponding numerical simulation. Flow direction is shown by the arrows (angle expressed relative to x). The dashed line is aligned with the injection port and provides a visual reference of the flow “centre line” along x .

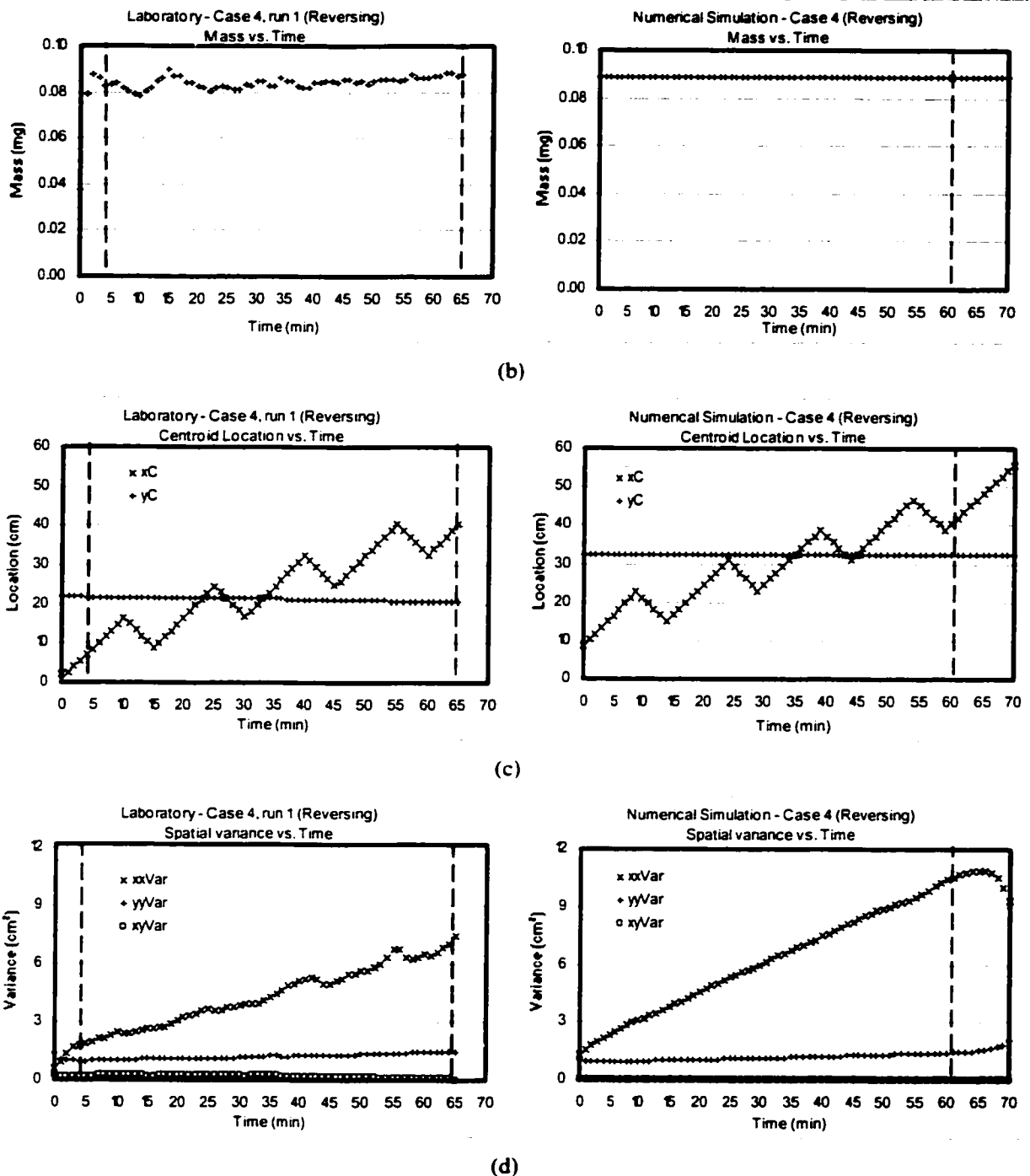


Figure 3-4. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 4 (reversing flow pattern parallel to longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

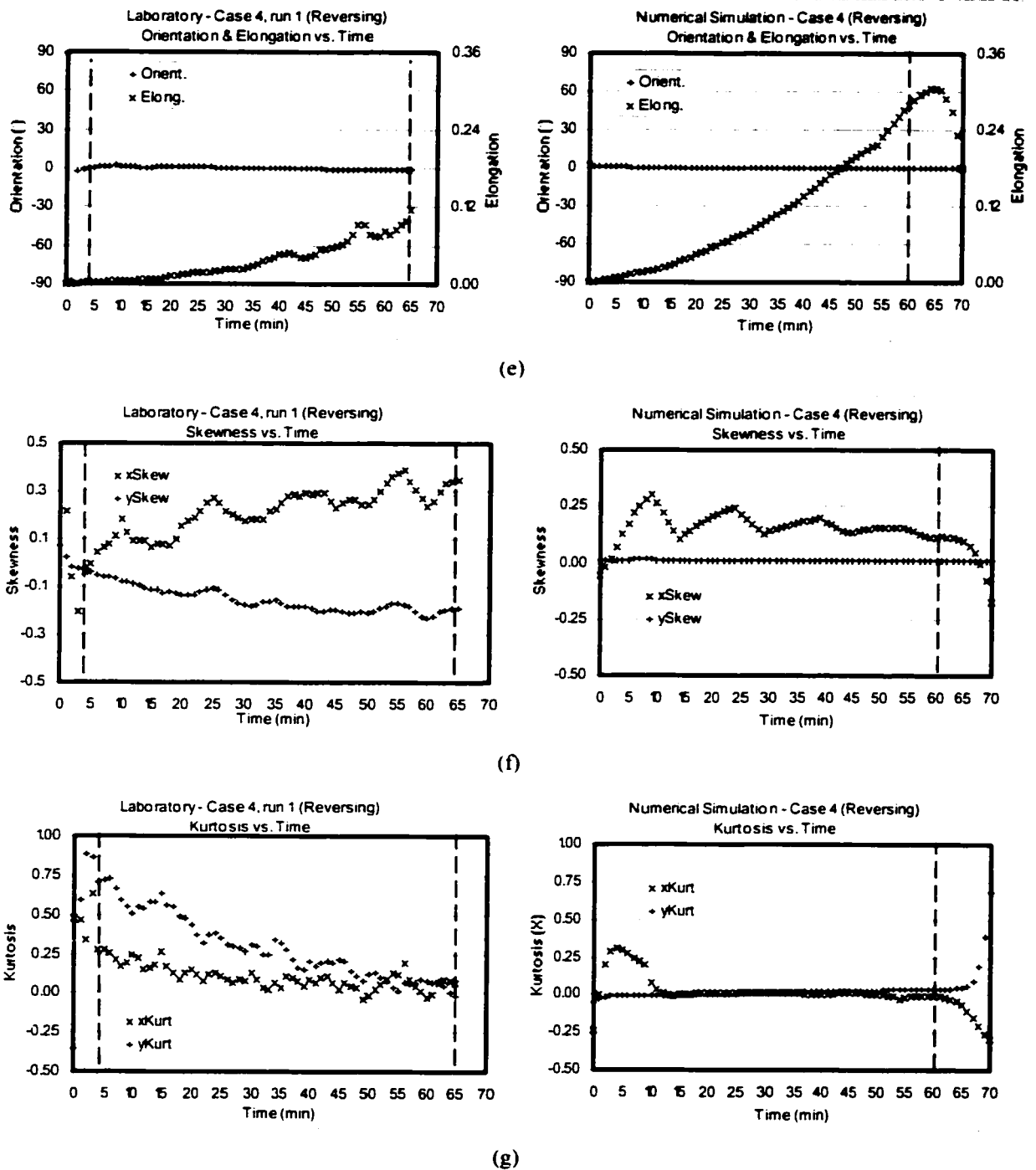


Figure 3-4. (continued)

Sample charts showing laboratory (left) and numerical (right) results for case 4 (reversing flow pattern parallel to longitudinal direction (x), $i = 0.1211$). Time series are shown for parameters derived from spatial moments of orders 1-4, including (b) total mass, (c) location of the centre of mass, (d) spatial variance, (e) orientation, (f) coefficient of skewness, and (g) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. The charts on the left were produced from image sequence in (a).

1.1 Mass balance

The total mass of dissolved solute (M_t) was estimated at each time step throughout the experiments, using concentrations derived from laboratory images or from numerical simulations. These estimates were then compared to the known injected mass $M_i = 0.084$ mg. The temporal evolution of solute mass was estimated by calculating spatial moments of order zero (M_{00}) of the concentration profiles obtained from (a) the calibrated image sequences and (b) from the transport simulations. M_t was estimated by first summing the elemental contribution of M_{00} from each pixel over the entire image for laboratory experiments; or from each grid cell of the simulation domain for numerical simulations. M_{00} was then adjusted to take into account the porosity ($n = 0.38$) and dimensions (volume) of each element, V_e . Typical results (using case 1) are shown in Table 3-1. The coefficient of variation ($CV = s_{\bar{M}_t} / \bar{M}_t$), calculated within the “acceptable” time interval (T) to quantify the variability around time-averaged $\bar{M}_t$, was between 1.3 and 4.1% (2.5% on average), and was less than 0.5% for numerical simulations.

Table 3-1. Total dissolved mass $\bar{M}_t$ estimated for case 1 (steady state flow, $i = 0.1211$) from laboratory experiments and from numerical simulations. Elemental volume V_e is obtained by multiplying the thickness of the flow cell (0.033 m) by the area of each pixel or grid cell ($2.21 \times 10^{-7} \text{ m}^2$ or $1.43 \times 10^{-5} \text{ m}^2$, respectively). Experimental results compare well with the known injected mass $M_i = 0.084$ mg.

Calculation method	V_e (m^3)	$\bar{M}_t$ (mg)	CV
1. Using M_{00} from image sequence (averaged over $t = 3\text{--}29$ min)	7.28×10^{-6}	0.087	2.6%
2. Using M_{00} from numerical simulation results	4.71×10^{-7}	0.089	0.1%
3. Calculated by MT3D for each simulation time (from output file)	4.71×10^{-7}	0.087	0.0%
4. Injected mass (known – 10 mL @ 8.40 mg L ⁻¹)	–	0.084	–

Typical plots were presented in part (b) of Figure 3-1 to Figure 3-4 to illustrate the evolution of M_t over time for the selected scenario. Note that these plots were useful in defining the “acceptable” time interval T (shown by the vertical dashed lines), when the plume was partially outside the visible portion of the flow cell. This range typically started at about $t = 2\text{--}3$ min, when the plume had completely cleared the (opaque)

injection port, and extended until the plume reached the right edge of the flow cell and the total mass started declining as the tracer exited through the outlet ports.

In general, M_t from laboratory experiments (0.085 mg on average) slightly underestimated total mass, but was generally consistent with the results from the numerical simulations (0.089 mg). The absence of a slope in the time plots of M_t and the small variability indicate that mass was preserved and correctly accounted for by the experimental methods.

The unusual behaviour of M_t vs. t for the reversing flow scenarios is worth noticing (case 4, run 1, shown on the left in Figure 3-4(b)). After a slight initial decrease, M_t increases sharply when flow is first reversed during the period $t = 10$ to 15 min. The mass then decreases during forward flow periods and increases during flow reversal events. This cyclic trend repeats as the experiment progresses, but with decreasing amplitude until it can no longer be recognized after 2-3 forward-reverse cycles. The successive periods of increasing and decreasing total mass are thought to be an artificial effect caused by a combination of two factors related to the experimental design: (1) the “wall effect” and (2) the local nature of the region of interest (ROI) used to calculate the spatial moments. These are illustrated in Figure 3-5 for the reversing flow scenario (case 4, run 1). The false shading (a banded grey scale lookup table) helps in defining the internal structure of the plume. At the end of the first “flow forward” interval ($t = 10$ min), the plume is seen to have developed a sort of protuberance (or “tail”) on the right, in the direction parallel to flow ($+x$). After the next flow sub-interval ($t = 15$ min), where flow took place in the opposite direction ($-x$), the plume appears to have lost its tail on the right, while the maximum concentration near the centroid has increased slightly (6.2 vs. 5.9 mg L⁻¹ at $t = 15$ and 10 min, respectively). During the second forward period ($t = 15$ to 25 min), the plume develops its right-hand tail again. This behaviour suggests that there exists a zone within the flow cell, presumably near the walls of the cell, where flow is faster than elsewhere in the porous medium. This “wall effect” distorts the plume and causes a portion of its mass to migrate outside of the oval ROI. The wall effect is a phenomenon commonly encountered in laboratory experiments involving a flowing fluid

through a porous medium packed in a container with smooth walls. Walls effects are discussed in Appendix F.

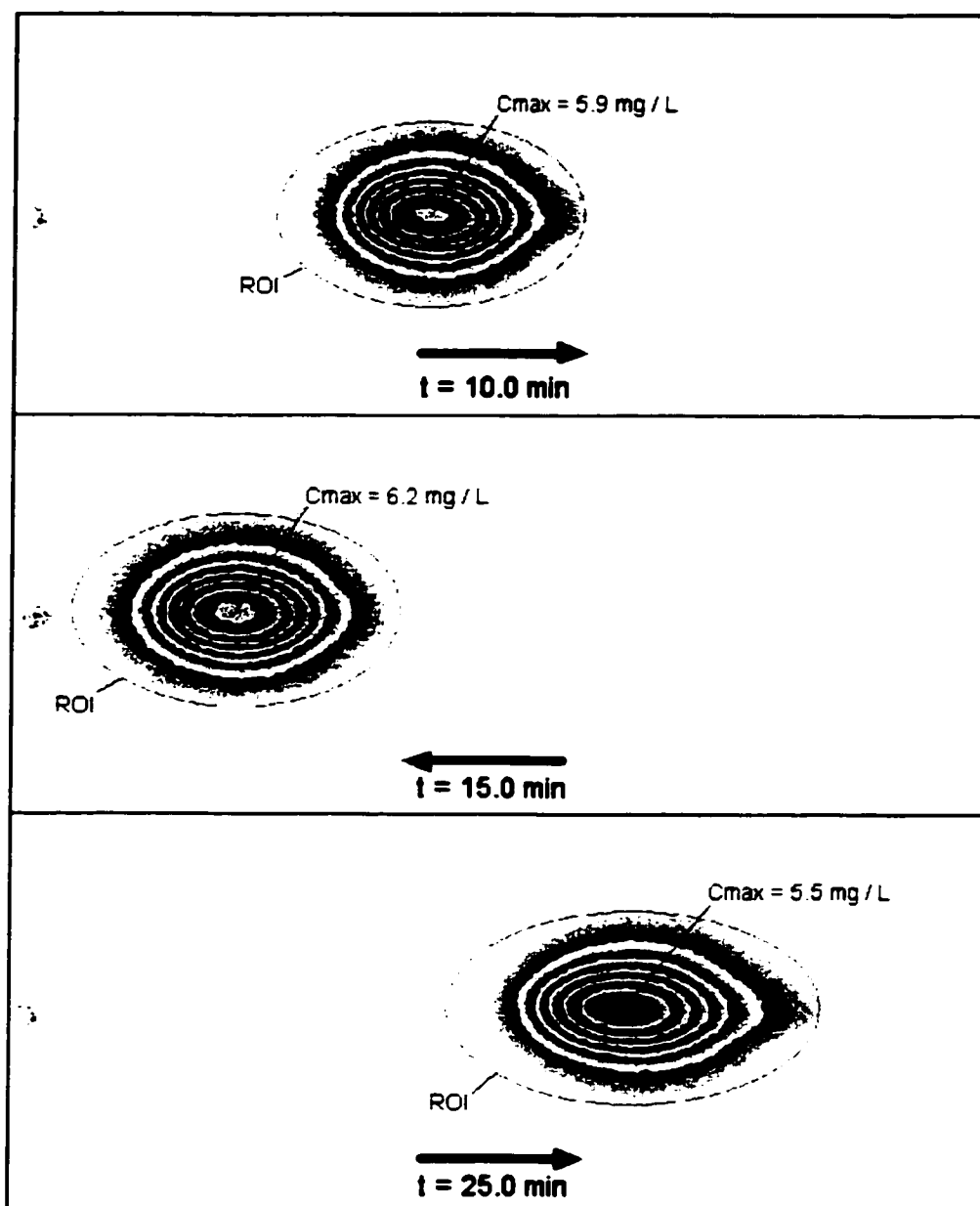


Figure 3-5. False-shaded image sequence from a reversing flow scenario (case 4, run 1). These images were acquired at times coinciding with the end of flow sub-intervals (flow direction is indicated by the arrows). The thin oval outlines the extent of the local region-of-interest (ROI) used to define the domain where moments were calculated. The banded colour scale was selected for illustrative purposes to enhance the internal structure of the concentration distribution. The sequence shows skewness (small "tail") developing to the right of the plume during forward motion and a contraction of the tail during reverse motion. This effect is thought to be caused by a "wall effect" – a zone of slightly higher permeability at the contact plane between the porous material and the wall of the flow cell. See Appendix F for details about wall effects.

While the wall effect appears to be responsible for the small distortion of the plume, the unusual “up and down” behaviour of the mass (M_t) described above is related to the procedure used by the plugin that calculates the spatial moments from the images. This plugin (see details in Appendix C) uses a local oval region-of-interest (ROI) (see Figure 3-5) to limit the domain over which the moment calculations are done, in order to reduce the effects of background noise outside the plume (these effects would dominate in the high-order moments). The plugin produces a ROI that moves and grows as the solute plume migrates and disperses. As shown in Figure 3-5, during the first forward flow interval (up to $t = 10$ min), M_t slowly decreases over time because a small fraction of the injected mass affected by the wall effects appears to be “lost” as it gradually migrates outside the oval ROI. When flow is reversed (at $t = 10$ min), the mass that was apparently lost during the previous interval of forward flow is gradually incorporated in the moment calculations as it re-enters the ROI and catches up with the plume. This behaviour becomes less apparent as the plume becomes increasingly diluted due to dispersion.

1.2 Centre of mass (centroid) and velocity

The coordinates of the plume centroid in the longitudinal (x) and transverse (y) directions were obtained from the normalized first-order spatial moments, $x_c = M_{10}/M_{00}$ and $y_c = M_{01}/M_{00}$, respectively (see Appendix C for details). The components of the plume velocity $\bar{v}_x$ and $\bar{v}_y$ were then obtained using:

$$\bar{v}_x = \frac{dx_c}{dt} \quad \text{and} \quad \bar{v}_y = \frac{dy_c}{dt} \quad (3.1)$$

and were used to calculate the magnitude of the plume’s average velocity $\|\mathbf{v}\|$, as outlined in Appendix D.

The evolution of x_c and y_c as a function of time was plotted in part (c) of Figure 3-1 to Figure 3-4. Note that spatial coordinates were measured from different reference points in images from laboratory experiments, compared to numerical simulations. As a result, the initial location of the centroid derived from images was $[x_c(t_0), y_c(t_0)] = (1.3, 21.8)$ cm, while for numerical simulations, its initial position was $[x_c(t_0), y_c(t_0)] = (8.9, 32.5)$ cm. As

expected, the flow pattern imposed on the system controlled the migration of the plume centroid. (shown in the above plots on the left and right, respectively) There was no apparent difference between the laboratory and the numerical results, suggesting that the numerical model adequately reproduced the advection process observed in the laboratory. Under time varying flow conditions, variations in the direction of the hydraulic gradient are clearly visible by the temporal evolution of y_c for the “zigzag” scenarios, or x_c for the reversing pattern.

1.3 Spatial variance

Second-order spatial moments were calculated as described in Appendix C. These are expressed relative to the centroid location, in order to account for the fact that the plume is not static in space. The central second moment (or spatial variance) thus provides a convenient way of estimating solute spreading around the plume’s centre of mass at any point in time. The time-rate of change of the spatial variance $d\sigma^2/dt$ will be used later to derive the coefficient of dispersion D , which is in turn used to calculate the macrodispersivity, A (see §2).

Part (d) of Figure 3-1 to Figure 3-4 show the temporal evolution of spatial variance along the longitudinal and transverse directions (σ_{xx}^2 and σ_{yy}^2 , respectively), as well as the spatial covariance (σ_{xy}^2). For laboratory results, variance estimates during the first few time steps (up to about $t = 2-3$ min) are inaccurate because the plume was partially hidden behind the injection port. Near the end of each experiment, as the plume approaches the outlet ports, distortions in the flow field cause a sharp decline in σ_{xx}^2 , accompanied by a sudden increase of σ_{yy}^2 . The limits of T (the “acceptable” time interval used to derive the dispersion coefficient) were determined from these visible effects on the time-plots of σ_{xx}^2 and/or σ_{yy}^2 . A pair of vertical dashed lines is shown on each plot from Figure 3-1 to Figure 3-4 to mark the interval’s starting and final times.

In all cases, σ_{xx}^2 , estimated from laboratory or numerical results, increases steadily with time as the plume spreads along the mean flow direction (parallel to x , i.e.

horizontally in the image sequences shown in Figure 3-1 to Figure 3-4). The rate of increase of σ_{xx}^2 is generally linear, with minor local deviations in the laboratory data due to errors related to the experimental setup and/or image processing and calibration procedure.

One noticeable trend in the laboratory results from the reversing flow scenarios (case 4) is the strong response of $\sigma_{xx}^2(t)$ to flow reversals (Figure 3-4(d)). This trend, which becomes increasingly important as the experiment progresses, has the slope of $\sigma_{xx}^2(t)$ steeper when flow is in the forward direction than when flow is reversed. The cyclical effect of flow reversals is hardly noticeable initially, but increases in intensity with each cycle until it becomes so strong that the slope $d\sigma_{xx}^2/dt$ can even become negative in the reverse direction. This negative slope indicates that the plume is actually shrinking during the late flow reversals (e.g. intervals $t = 40-45$ and $55-60$ min). This behaviour, which is not seen in the numerical simulation results, may be, at least partly, related to the wall effect discussed above. Since the main focus of this study is dispersion, which is described by the time derivative of the second moment, in-depth discussion of the time derivative behaviour will be the topic of an entire section at the end of this chapter (see §2).

The spatial covariance (σ_{xy}^2) gives an indication of the degree to which an elongated plume is misaligned with the axes. In all flow scenarios, the covariance was found to be very small (close to zero), indicating that the plumes were aligned with the axes or had no direction of elongation. However, changes in the flow direction induced slight distortions in the shape of the solute plume (e.g. zigzag cases 2 and 3, as shown in Figure 3-2(d) and Figure 3-3(d), respectively). Such distortions appear to be closely linked to the evolution of the vertical plume location (y_c vs. t), shown for these two cases in Figure 3-2(c) and Figure 3-3(c), respectively.

1.4 Elongation and orientation

Orientation is the angle between the longest axis of the plume and the positive x direction. Elongation, on the other hand, is a shape descriptor derived from the second-order moment (spatial variance) as the ratio between the plume's longest and shortest axes, irrespective of the plume orientation. Part (e) in Figure 3-1 to Figure 3-4 present the evolution over time of plume orientation and elongation. As expected, plume elongation increases over time as the contrast between the longitudinal and transverse dimensions gradually becomes larger due to the ever-increasing influence of hydrodynamic dispersion in the longitudinal direction relative to transverse dispersion. For the steady state scenario (e.g. Figure 3-1(e)), plume elongation increases slightly faster in numerical simulations compared to the laboratory results, and reaches the same maximum of about 0.06 two minutes earlier.

Time-variations in the direction of the hydraulic gradient have a significant influence on plume elongation. For the flow scenario with $\pm 30^\circ$ variations in the mean gradient direction (Figure 3-2(e)), the maximum elongation is smaller than for the steady state case (0.036 vs. 0.06). A similar trend is observed for the $\pm 60^\circ$ case (Figure 3-3(e)), where the elongation is even lower (up to about 0.015) for most of the duration of the experiment. This can be explained by the fact that each time the mean flow direction is rotated, the new hydraulic gradient acts on the plume at an oblique angle; this tends to slow the growth of the plume's longest axis, while enhancing its dimensions along the short axis. In other words, shifting the direction of the hydraulic gradient causes the shape of the plume to become bulkier. The temporal evolution of plume elongation derived from simulation results clearly shows the impact of changing the flow direction (see the charts on the right of Figure 3-2(e) and Figure 3-3(e)).

For the reversing flow scenario (case 4, shown in Figure 3-4(e)), the rate of increase of the plume elongation is almost three times slower in the laboratory experiment than in the numerical simulation. In the numerical simulations, the effect of reversing the flow direction has little influence on plume elongation compared to the steady state case. For instance, plume elongation at $t = 27$ min is only slightly smaller (0.061 vs. 0.069) for the

reversing case than for the steady case. Conversely, the laboratory-derived estimate of plume elongation suggests that the plume can actually shrink during flow reversal periods (e.g. for the intervals $t = 40\text{--}45$ min and $t = 55\text{--}60$ min). This phenomenon might be due in part to the wall effects discussed previously. However, the large discrepancy between laboratory and numerical results also suggests that the mathematical model does not accurately capture the effect of reversing flow on solute dispersion.

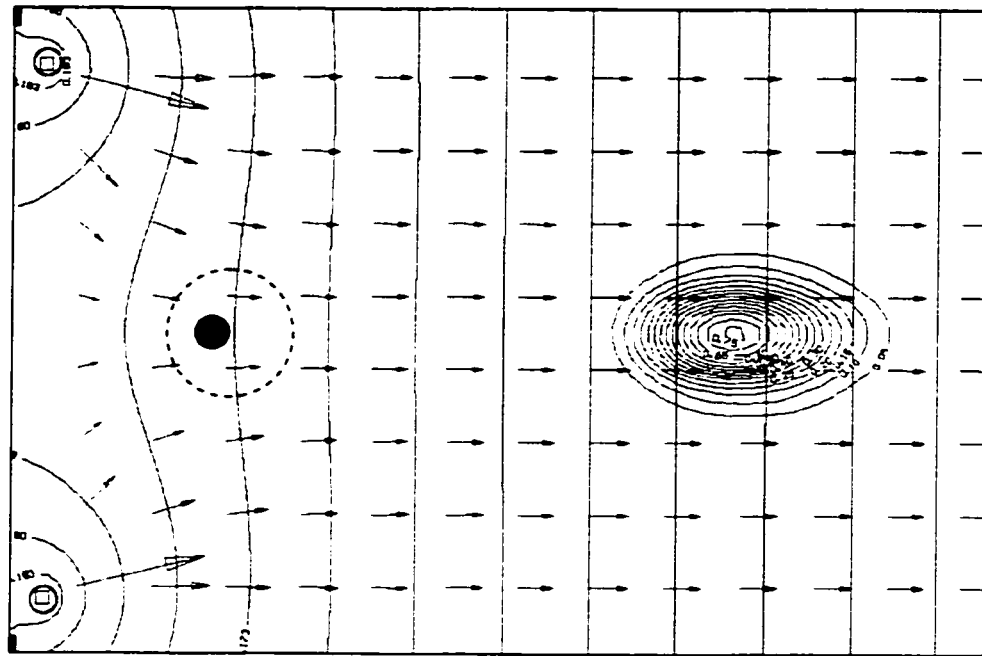
Plume orientation is significantly influenced by deviations in the flow direction from the longitudinal (x) axis, unlike for the steady state and reversing flow scenarios where plume orientation remained close to zero throughout the duration of the experiments. A change in the gradient direction, however, causes the plume to rotate such that its longest axis gradually reorients itself parallel to the new flow direction. Plume orientation from the zigzag scenarios was never allowed to reach the direction imposed by the gradient following a change. This is explained by the relatively brief time sub-intervals of temporarily steady flow conditions. The Δt values were specified to ensure that the plume remained uninfluenced by the inlet/outlet ports surrounding the flow domain, and were thus limited by the mean hydraulic gradient and by the limited physical dimensions of the laboratory model. Consequently, each sub-interval was too brief to allow the plume's orientation to parallel the new flow direction (see results for cases 2 and 3, shown in Figure 3-2(d) and Figure 3-3(d), respectively).

1.5 Coefficient of skewness

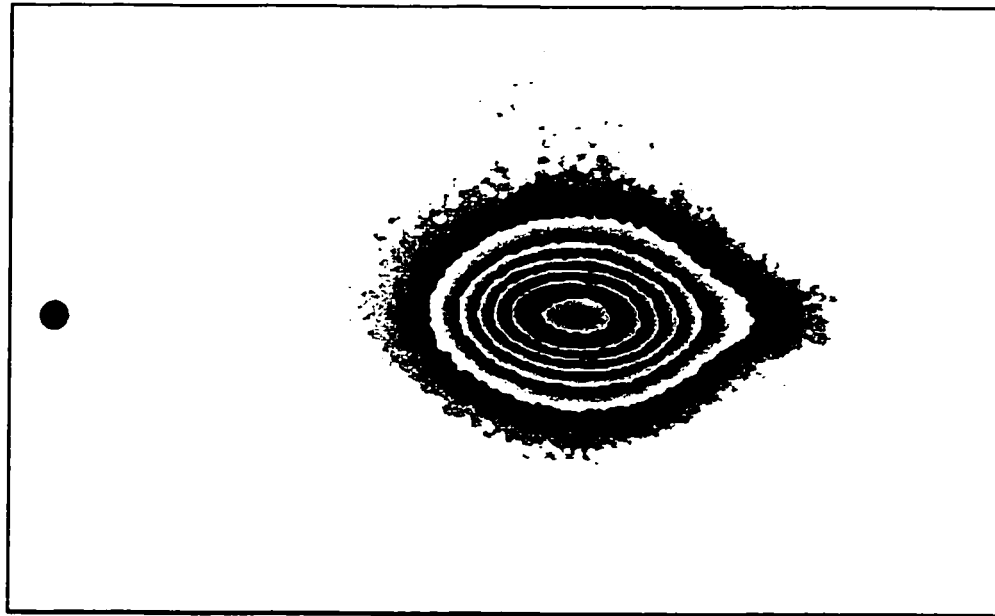
Plume symmetry in the direction of the axes can be assessed from the coefficient of skewness, which is derived from the third-order spatial moments. $SK = 0$ for a normal distribution. Plots showing the evolution of SK_x and SK_y vs. t are shown for cases 1 to 4 in part (f) of Figure 3-1 to Figure 3-4, respectively. Note that during the initial time steps (up to about $t = 3$ min), the unusually high estimates of SK_x calculated from laboratory experiments are not representative of the plume shape because a significant portion of the plume was obstructed by the injection port.

The initial shape of the plume for all flow scenarios is nearly symmetrical, with $SK_x \approx SK_y \approx 0$ for both laboratory and numerical results. SK_x for all flow scenarios investigated

increases rapidly as the migrating plume becomes skewed towards the right. This can be explained by the convergent nature of the flow field near the injection port, as illustrated in Figure 3-6. In order to produce a symmetrical initial plume, the extents of which are shown by the dashed circle in Figure 3-6(a), flow inside the laboratory model was interrupted during tracer injection. When flow resumed following tracer injection ($t = 0$), the plume began migrating (by advection) and spreading (due to dispersion) along the direction of flow. While the left portion of the plume was influenced by the lower velocities induced by the zone of converging flow, flow velocities at the front (right edge) of the tracer pulse were slightly larger than at the rear (left). Consequently, the differential velocities at the right and left of the plume caused it to become increasingly asymmetrical towards the right at early times. The increase in SK_x is relatively fast at the beginning of each experiment, but gradually slows down as the influence on the plume of the zone of convergence diminishes; SK_x reaches a plateau when the plume has migrated far away from the inlet/outlet ports ($t \approx 10$ min for flow scenarios with a gradient of 0.1211).



(a)



(b)

Figure 3-6. Solute plume ($t = 10$ min) from the steady state flow scenario ($i = 0.1211$) showing the asymmetrical concentration distribution (skewed towards the right). Results from the numerical simulation are shown in (a), including solute concentration contours (C/C_0), equipotential lines and seepage velocities (arrows). Notice how the arrows to the left of the injection port (black circle) converge slightly. The initial ($t = 0$) extents of the tracer pulse are shown using the dotted line in (a). The image shown in (b) was taken from the laboratory experiment case 1, run 1. For illustrative purposes, a banded greyscale lookup table (LUT) was used to enhance the contrasts in tracer concentrations.

The asymmetry is generally slightly more pronounced for laboratory experiments compared to numerical simulations, and does not appear to reach a plateau. This discrepancy might be explained by the wall effects caused by the thin zone of increased permeability at the flow cell's front and back panels. This phenomenon will be discussed in Appendix F. Wall effects cause groundwater to flow slightly faster at the interface between the porous medium and the flat walls of the experimental model. The dissolved tracer is therefore susceptible to respond more quickly to flow transience in those zones than within the interior of the porous medium, where the influence of the cell's walls has been damped out. It is possible to discriminate between the wall effect and physical effects of flow transience by studying the temporal behaviour of SK_x for scenarios where hydraulic gradients are periodically reversed.

In the case of the reversing flow scenarios (see e.g. Figure 3-4(f)), SK_x decreases during each flow reversal period, and increases during the "flow forward" sub-intervals. This cyclical behaviour is apparent in both the laboratory and the numerical results. In the laboratory model, the overall trend is for SK_x to continue to increase over time, while in the numerical model SK_x the cycles are superimposed upon a plateau and tend to decrease in amplitude over time. In other words, the succession of flow reversals causes the "numerical" plume to gradually show a constant degree of asymmetry over time. The increasing overall trend in the laboratory compared to the numerical results may be explained in part by the wall effects, which obviously only affect the laboratory results. The cyclical behaviour may be explained by the converging flow field shown in Figure 3-6. As the plume advances in the forward direction the flow field constriction increases the skewness, but when it advances in the reverse direction, the fanning-out flow field produces smaller velocities on the left than the right, thereby reducing the right-hand side skewness.

In the transverse direction, SK_y becomes increasingly negative over time for laboratory results, indicating that the vertical asymmetry develops upwards (-y direction). This behaviour is in line with the slight upward movement experienced by the plume's centre of mass over time (see Figure 3-4(c) for a plot of y_c vs. t). The upward movement is likely due to the hydraulic gradient being somewhat off from the intended left-right flow

axis. The development over time of the plume's "tail" (monitored from SK_y vs. t) in the same direction suggests that the wall effect is significant, since its evolution is faster than that of y_c vs. t . This is consistent with the theory that the increased porosity near the smooth walls enhances permeability [e.g. *Freeze and Cherry*, 1979]. This "fast" zone causes the effects of changing hydraulic gradients to appear exaggerated. On the other hand, the plume's vertical symmetry is preserved throughout the numerical simulation, with $SK_y \approx 0$.

1.6 Kurtosis

Derived from the fourth-order moments, the kurtosis (KR) provides a description of the degree of peakedness (or flatness) of a distribution. For normally distributed data, $KR \approx 0$, while a positive or negative kurtosis indicates a distribution that is either more peaked or flatter than normal, respectively. Part (g) of Figure 3-1 to Figure 3-4 illustrate the evolution over time of the x and y components of kurtosis for cases 1 to 4.

For all flow scenarios, the plume's longitudinal kurtosis (KR_x) increases rapidly at the first time steps and then decreases progressively towards zero. This is the result of small imperfections in the shape of the freshly injected plume which get smoothed out by dispersion as time progresses.

In the transverse (y) direction, laboratory and numerical results are noticeably different. During laboratory experiments, KR_y increases rapidly until it reaches a maximum of about 0.8, and then decreases over time whereas it remains near zero for the numerical simulations. Comparing the steady state scenario (Figure 3-1(g)) with the non-steady cases (Figure 3-2(g), Figure 3-3(g) and Figure 3-4(g)) suggests that flow transience apparently speeds up this "flattening out" process. Interestingly, reversing the hydraulic gradient in the longitudinal direction appears to reverse the trend in KR_y , which decreases during forward flow intervals but increases (more slowly) when flow is reversed (e.g. case 4 in Figure 3-4(g)). This behaviour is surprising because changes in the flow direction are imposed in the direction perpendicular to that of KR_y . One possible explanation for this phenomenon is that it results from a combination of two factors that distinguish the laboratory conditions from the "ideal" conditions specified during the

numerical simulations. First, the slight upward centroid migration due to an imperfect flow field artificially enhances transverse mixing. Second, the wall effects discussed previously (see §1.1) amplify the flattening of the plume during forward flow intervals, while apparently undoing this (in part) when flow is reversed: the “fast” portion of the plume resulting from the wall effects is gradually superimposed back onto the rest of the plume.

2 MACRODISPERSIVITY

Macrodispersivity (A_{ij}) was used to describe the overall dispersive behaviour of solute plumes for the various flow scenarios. It was calculated as:

$$A_{ij} = \frac{1}{\|\mathbf{v}\|} D_{ij} \quad (3.2)$$

where $\|\mathbf{v}\|$ is the magnitude of the plume’s bulk velocity and D_{ij} is the macroscopic dispersion coefficient tensor. These were estimated from the time rate of change of the plume’s moment-derived centroid and spatial variance, respectively. Appendix D discusses the methodology used to calculate A_{ij} , which, as will be shown in the following sections, can be quite different depending on the selection of the time interval(s) of the time derivative(s) used to calculate $\|\mathbf{v}\|$ and D_{ij} . Of the four methods that were identified (*Global*, *Stepwise*, *Absolute Stepwise* and *Finite Difference*), only the *Absolute Stepwise* method was found to be significantly different from the others. Since the other methods were found to be generally equivalent (i.e. all three methods produced similar results for a given scenario), data computed from the *Stepwise* and *Finite Difference* methods are not presented here; instead, the *Global* method was selected as being representative of these two methods. Consequently, experimental results for all scenarios are presented in parallel, including those obtained from both the *Global* and from the *Absolute Stepwise* methods. The *Global* method estimates the time derivative from the regression line of the time-plots, whereas the *Absolute Stepwise* method takes the average of the absolute values of the time derivatives of each time interval. The latter method is of particular relevance when the flow direction varies and velocities can be positive or negative

depending on the time interval. In lay terms, the *Global* method gives estimates of the time derivatives that ignore the small time scale fluctuations, as though the observer simply saw the first and last snapshots of the plume, without knowledge of the time behaviour of the flow field in between. Conversely, the *Absolute Stepwise* method takes into account each time interval irrespective of the actual flow direction, as though the intricate and sometimes reversing flow path was unravelled into a single unidirectional flow path.

2.1 Confidence intervals

The uncertainty in the experimental results from a given scenario was estimated by calculating the 95% confidence intervals (*CI*). The *CI* for a given parameter can be illustrated graphically using vertical error bars on graphs, or in the form of limits expressed as $\pm CI$, given by:

$$CI = t_{\frac{(1-\alpha)}{2}, DF} \left(\frac{SE}{\eta} \right) \quad (3.3)$$

where t is the Student statistic obtained from standard tables, α is the level of significance, $(1 - \alpha)$ is the confidence level (95% in this case), DF is the number of degrees of freedom associated with the parameters in question, SE is the standard error of the statistic being evaluated; and η is the number of replicates included in the statistic of interest. A confidence level of 95% was used in this study ($\alpha = 0.05$).

The procedure used to estimate SE and DF is described in detail in Appendix D. Several factors were considered in estimating the experimental error, including:

1. The standard error on the slope of the regression line: for the *Global* method, there was a single SE estimate with a relatively large number of degrees of freedom. Conversely, for the *Absolute Stepwise* method, there were several SE estimates, one for each of M time intervals, and each estimate with few degrees of freedom. For those calculations that involved multiple regression intervals, the combined SE for each experimental run was derived from a time-weighted

average of M estimates of SE , using the duration of each sub-interval as the weighting factor.

2. Precision error: For the laboratory experiments, estimates of $\bar{v}_i$ and D_{ij} for a given scenario were pooled over a number of replicate runs ($\eta = 2$ to 6). The SE from each run were combined for each parameter using the number of degrees of freedom from each run as the weighting factor. On the other hand, numerical results were derived from simulations where all parameters were specified deterministically, and precision error was therefore not applicable.
3. Propagation of error: The uncertainties, expressed as SE , associated with $\bar{v}_i$ and D_{ij} were combined when calculating derived quantities ($\|\mathbf{v}\|$, A_i , and the ratio A_x/A_y) by applying the superposition-of-error theorem [Leaver and Thomas, 1974] on the means averaged over multiple runs for a given scenario.

2.2 Results

The charts summarizing the laboratory results are presented in Figure 3-7 to Figure 3-9, while results from numerical simulations are shown in Figure 3-10 to Figure 3-12. In the charts below, the longitudinal (x) and transverse (y) components are presented side by side for each flow scenario, with the lower-gradient scenarios (cases 1-4) shown on the left, and the higher-gradient scenarios (cases 5-8) on the right. Recall from the previous chapter the four distinct flow patterns investigated, namely the steady state, low- and high-angle zigzag (30° and 60° , respectively), and reversing scenarios.

The laboratory data discussed in this section were averaged as described in Appendix D over a number of replicate experiments (with η varying from 2 to 6 depending on the scenario). Detailed results from all runs are shown individually in Appendix F.

Longitudinal (A_x) and transverse (A_y) macrodispersivities obtained from laboratory results using the *Global* and *Absolute Stepwise* methods are shown in Figure 3-7(a) and Figure 3-7(b), respectively. For each flow scenario, the ratio A_y/A_x is presented in Figure 3-8, while Figure 3-9 shows the magnitude of the velocity vector $\|\mathbf{v}\|$.

A comparison of macrodispersivity between steady and transient scenarios shows a number of significant trends. Looking at the laboratory results (*Global* method) for the lower gradient scenarios (left-half of Figure 3-7(a)), we see that shifting the flow direction causes A_x to decrease proportionally to the rotation angle, while A_y increases. In the reversing scenario, on the other hand, A_x and A_y are both significantly larger than in the steady state case. Results obtained during the experiments where the hydraulic gradient was higher (plotted in the right-half of Figure 3-7(a)) are identical for a given flow pattern. This is true of both A_x and A_y . This suggests that the two velocities selected were within the bounds of Fickian behaviour for the dispersion process, and that the differences in macrodispersivities between the scenarios are due to transience in the flow direction, not in the magnitude of the velocity vector.

A similar trend is observed from the *Absolute Stepwise* method, whose results are shown in Figure 3-7(b). Here again, rotating the flow direction causes A_x to decrease and A_y to increase in proportion to the rotation angle ($A_x^{(1)} > A_x^{(2)} > A_x^{(3)}$, and $A_y^{(1)} < A_y^{(2)} < A_y^{(3)}$, where the subscripts refer to the steady state, zigzag30° and zigzag60° patterns, respectively). Note, however, that the relative differences between scenarios appear to be amplified using this calculation method (A_x decreases faster and A_y increases more slowly as the rotation angle increases). The most striking difference between the two methods, however, appears to be in the reversing scenario. Surprisingly, A_x is smaller for the reversing scenario compared with the steady state case, while no significant difference is observed in the case of A_y . As before for the *Global* results, no difference could be observed between the two hydraulic gradients (comparing A_x or A_y for a given flow pattern).

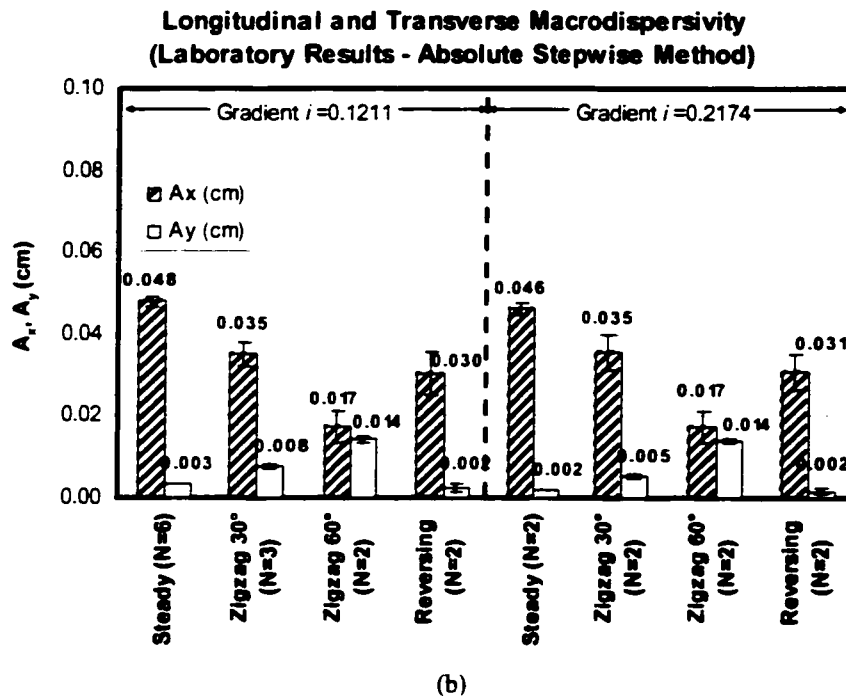
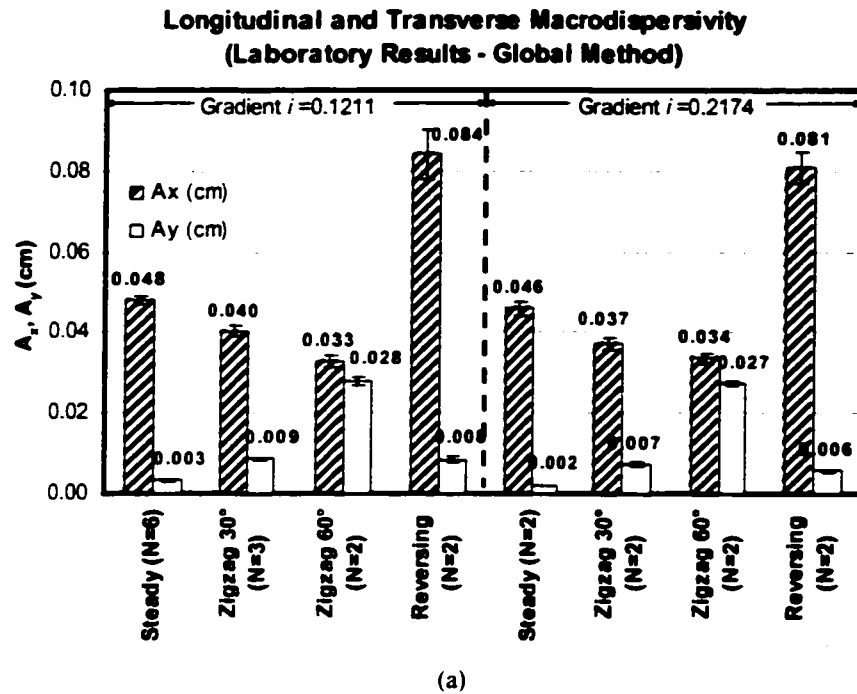


Figure 3-7. Longitudinal and transverse macrodispersivity (A_x and A_y , respectively) estimated from laboratory data using the *Global* (a) and *Absolute Stepwise* (b) calculation methods. For each of the eight flow scenarios, data were averaged over 2-6 replicate runs; the number of replicates for each case is shown as N . Error bars indicate the 95% confidence interval.

The relative impact of flow transience on longitudinal vs. transverse macrodispersivity can best be assessed by plotting the ratio A_y/A_x for each scenario. This was done in Figure 3-8, where results obtained for all scenarios using the *Global* and *Absolute Stepwise* methods are compared. First, we see that A_y/A_x increases rapidly with increasing rotation angle in flow direction, compared with the steady state case. This is an intuitive result since the sideways motion is expected to produce more lateral mixing. Also not surprising is the fact that the reversing flow scenario had only a small influence on the dispersivity ratio, marginally increasing the ratio from 0.07 to 0.10. In this case, there is no lateral water movement to increase lateral mixing. The calculation method has no significant impact on the ratio A_y/A_x , suggesting that the averaging technique did not affect the aspect ratio of the plume.

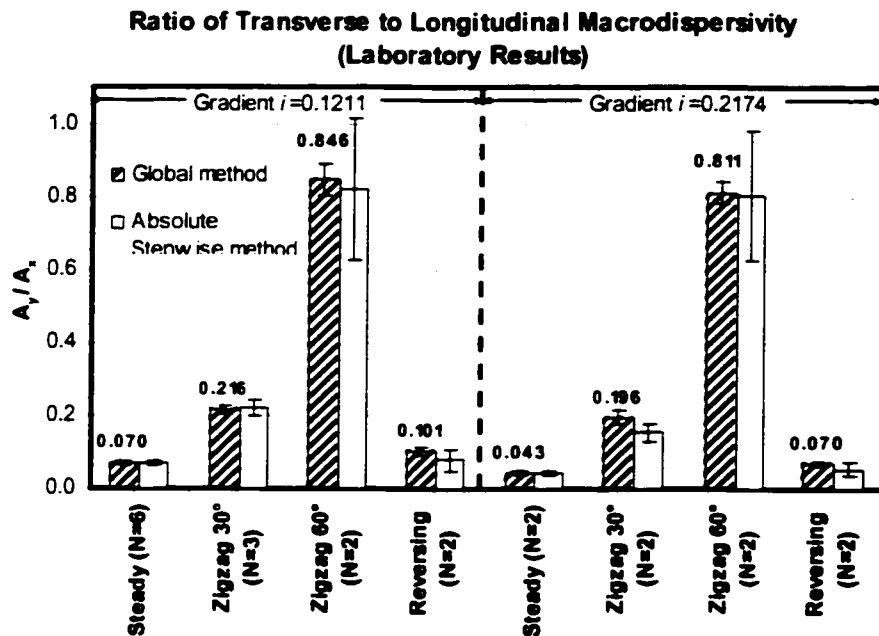


Figure 3-8. Ratio of transverse to longitudinal macrodispersivity (A_y/A_x), from results estimated from laboratory data using the *Global* and *Absolute Stepwise* calculation methods. For each of the eight flow scenarios, data were averaged over 2-6 replicate runs; the number of replicates for each case is shown as N . Error bars indicate the 95% confidence interval.

The magnitude of the velocity vector is plotted for all cases in Figure 3-9, where the low- and high-gradient scenarios are shown on the left and right, respectively. Results obtained using both the *Global* and *Absolute Stepwise* methods are plotted side-by-side for each scenario. Unsurprisingly, a comparison of $\|\mathbf{v}\|$ between the two gradients reveals that velocity for any of the flow patterns is directly proportional to the magnitude of the hydraulic gradient. The calculation method has a significant influence on $\|\mathbf{v}\|$; the velocities estimated using the *Global* method appear to be smaller for transient cases compared to the steady state scenario because the *Global* method essentially averages the velocities in the x axis direction. The *Absolute Stepwise* method, on the other hand, produces $\|\mathbf{v}\|$ estimates that are identical for all cases within a given set, where the hydraulic gradient remains constant. This is so because this method ignores the gradient direction, and only looks at the magnitude, which did not change in the course of the experiments.

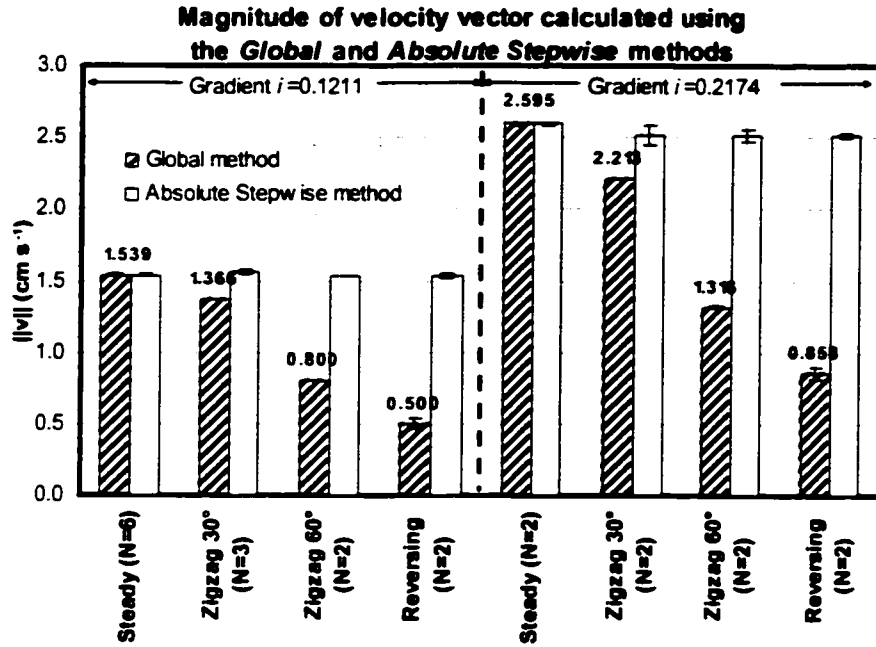


Figure 3-9. Magnitude of the velocity vector $\|v\|$ from laboratory results, estimated using the *Global* and *Absolute Stepwise* calculation methods. For each of the eight flow scenarios, data were averaged over 2-6 replicate runs; the number of replicates for each case is shown as N . Error bars indicate the 95% confidence interval.

Results from numerical simulations are presented graphically in the following figures using the same format as the preceding section. Longitudinal (A_x) and transverse (A_y) macrodispersivities obtained from laboratory results using the *Global* and *Absolute Stepwise* methods are shown in Figure 3-10(a) and Figure 3-10(b), respectively. For each flow scenario, the ratio A_y/A_x is presented in Figure 3-11, while Figure 3-12 shows the magnitude of the velocity vector $\|v\|$.

The general trends observed from the laboratory results with respect to A_x and A_y can also be seen from the results obtained from the numerical simulations. As before, hydraulic gradient rotation results in lowering A_x while A_y increases. Also, reversing the flow direction causes A_x and A_y to increase when estimated using the *Global* method, while both quantities remain unchanged compared to the steady case if they are estimated using the *Absolute Stepwise* method. Note also that the discrepancy between the high- and low-gradient cases for a given flow pattern can be quite significant. For example, the

increase in A_x is not as large for high-gradient reversing case, while A_y increases by a larger amount compared to the steady state scenario. This is true for both methods (Figure 3-10(a) and Figure 3-10(b), respectively).

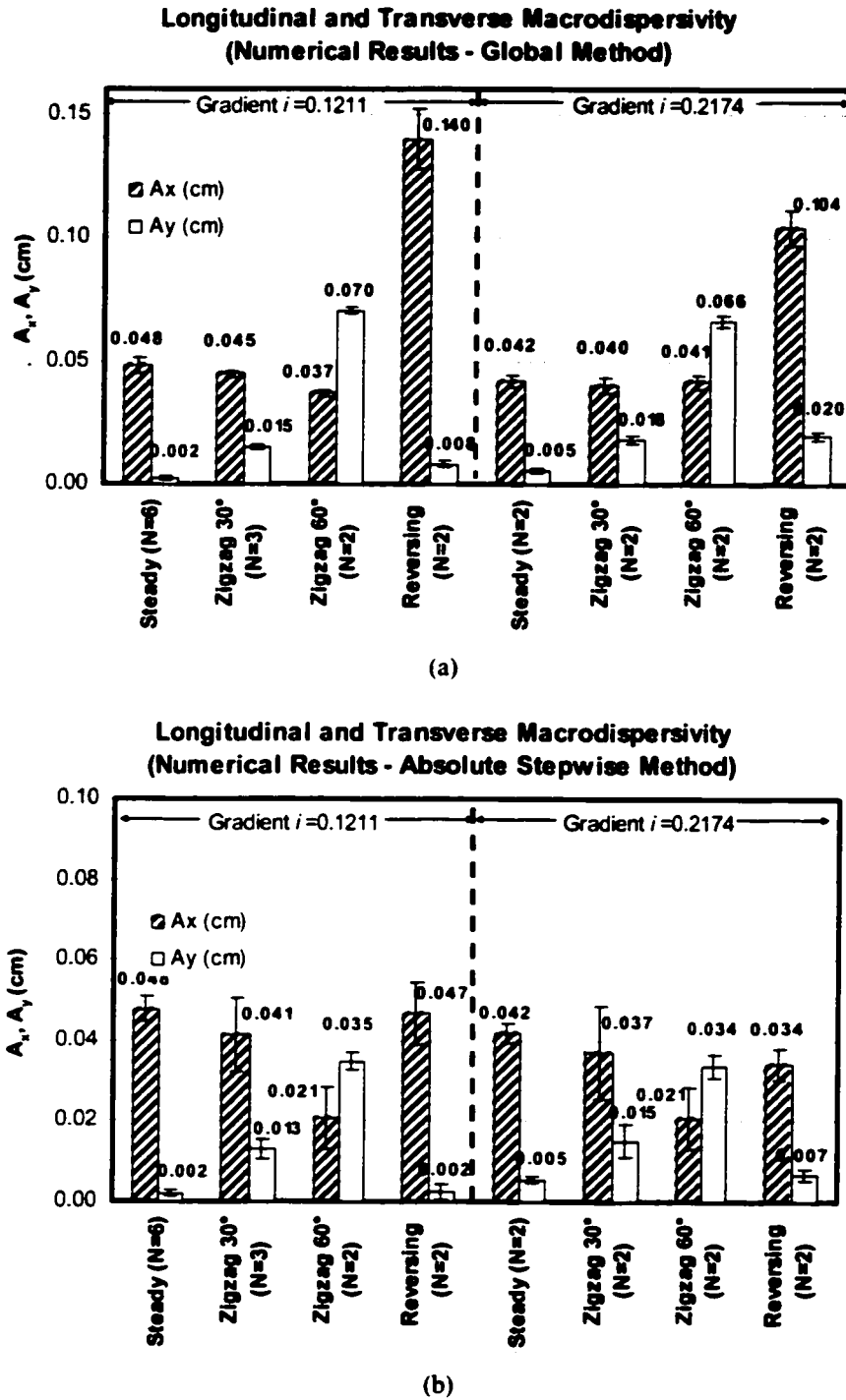


Figure 3-10. Longitudinal and transverse macrodispersivity (A_x and A_y , respectively) estimated from numerical simulations calculated using the *Global* (a) and *Absolute Stepwise* (b) methods. Error bars indicate the 95% confidence interval.

Comparing the ratio A_y/A_x for the numerical simulations from the *Global* and *Absolute Stepwise* methods (Figure 3-11) shows that, while no significant differences are seen between the methods, the ratio calculated using the latter is marginally lower. As in the laboratory results, shifting the direction of the gradient causes A_y to increase relative to A_x , as seen from the increase in the ratio A_y/A_x . One noticeable difference, however, is that the ratio increases by a larger proportion in numerical results compared to the laboratory results for a given scenario. This observation will be discussed in greater detail shortly. Also, the differences between the steady and reversing scenarios are small for a given gradient magnitude.

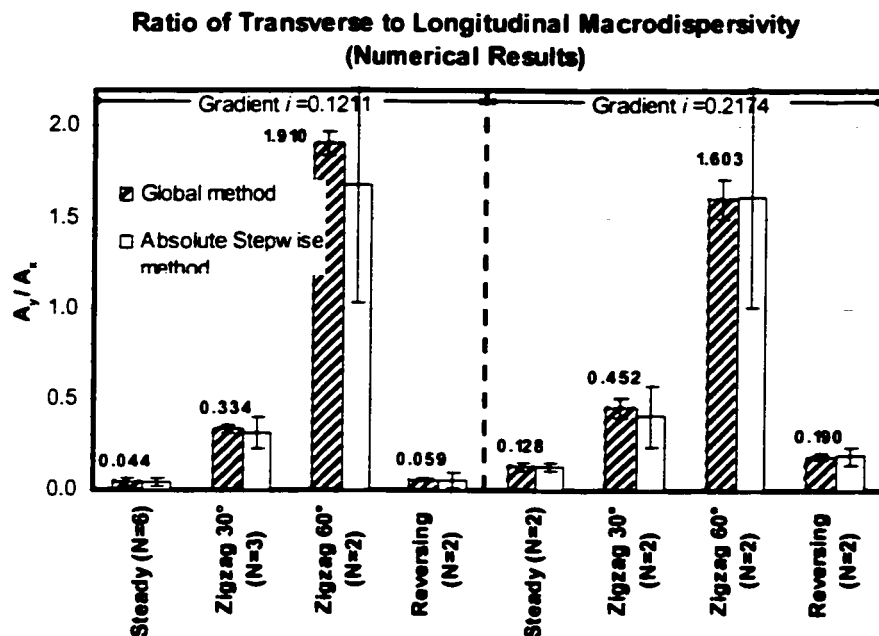


Figure 3-11. Ratio of transverse to longitudinal macrodispersivity (A_y/A_x) from numerical simulation results, estimated using the *Global* and *Absolute Stepwise* calculation methods. Error bars indicate the 95% confidence interval.

Figure 3-12 plots the magnitude of the velocity vector $\|\mathbf{v}\|$ estimated from numerical simulations using both calculation methods. Results are almost identical to those from the laboratory experiments (Figure 3-9). Only marginal differences are observed, namely the

slightly higher velocities for results from cases of higher gradient, and a small increase in uncertainty in some cases.

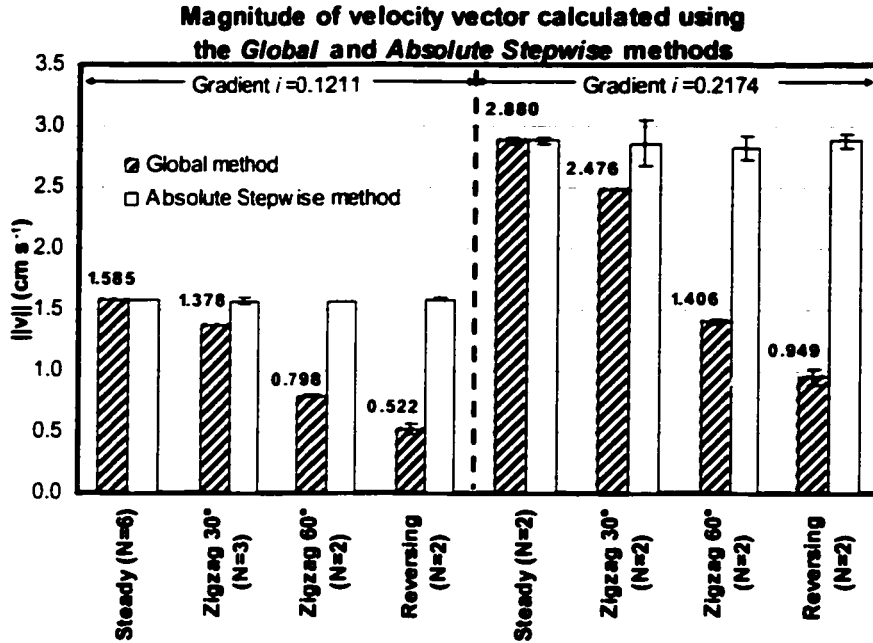


Figure 3-12. Magnitude of the velocity vector $\|v\|$ from numerical simulations, estimated from the *Global* and *Absolute Stepwise* calculation methods. Error bars indicate the 95% confidence interval.

2.3 Effect of hydraulic gradient magnitude

The influence on macrodispersion of the hydraulic gradient magnitude can be assessed by comparing each parameter (A_x , A_y , A_y/A_x , or $\|v\|$) for each flow scenario. Table 3-2 compares the magnitude of the velocity $\|v\|$ for the high hydraulic gradient ($i_2 = 0.22$) with $\|v\|$ for the low hydraulic gradient ($i_1 = 0.12$). The comparison was made by calculating the ratio $R_{\|v\|} = \|v_1\| / \|v_2\|$ for each scenario and calculation method. $R_{\|v\|}$ varied from 1.61 to 1.72 in the laboratory and from 1.76 to 1.82 in the simulations. This is very close to the ratio of the gradients, $R_i = i_2 / i_1 = 0.22 / 0.12 = 1.80$. This is true for laboratory and numerical results, regardless of the method used to calculate $\|v\|$. The

small discrepancy between laboratory and numerical results ($R_{\|\mathbf{v}\|}$ is slightly smaller in the former) is likely due to an inaccurate measurement of the gradient and/or conductivity of the laboratory flow cell.

Table 3-2. Comparison of the magnitude of the flow velocity vector $\|\mathbf{v}\|$ between high and low hydraulic gradients ($i_2 = 0.2174$ and $i_1 = 0.1211$, respectively). The ratio $R_{\|\mathbf{v}\|}$ is shown for each pair of scenarios where the configuration of the flow direction was the same (e.g. case 5 vs. case 1, case 6 vs. case 2, and so on).

Flow scenario	Gradient magnitude, i		$R_{\ \mathbf{v}\ } = \ \mathbf{v}_1\ / \ \mathbf{v}_2\ $			
			Laboratory experiments		Numerical simulations	
	$i_1 = 0.12$	$i_2 = 0.22$	Global	Absolute Stepwise	Global	Absolute Stepwise
Steady state	case 1	case 5	1.69	1.69	1.82	1.82
Zig-zag 30°	case 2	case 6	1.62	1.61	1.80	1.82
Zig-zag 60°	case 3	case 7	1.65	1.63	1.76	1.81
Reversing	case 4	case 8	1.72	1.64	1.82	1.82

Each component of macrodispersivity A_i can be compared using similar ratios. Referring to Figure 3-7 and to Figure 3-10, showing plots of A_x and A_y for each scenario the ratios were calculated and are presented in Table 3-3 and Table 3-4, respectively.

Longitudinal macrodispersivity A_x (Table 3-3) is essentially independent of the hydraulic gradient magnitude, as indicated by the ratio $R_{A_x} \approx 1$. Note that for numerical results, A_x is marginally lower than expected (i.e. slightly less than one), especially for the reversing flow scenarios, where R_{A_x} values are as low as 0.73.

Table 3-3. Comparison of the longitudinal macrodispersivity A_x between high and low hydraulic gradients ($i_2 = 0.22$ and $i_1 = 0.12$, respectively). The ratio R_{A_x} is shown for each pair of scenarios where the configuration of the flow direction was the same (e.g. case 5 vs. case 1, case 6 vs. case 2, and so on). A value of one indicates that results from both gradients produce identical results.

Flow scenario	Gradient magnitude, i		$R_{A_x} = A_{x_2} / A_{x_1}$			
			Laboratory experiments		Numerical simulations	
	$i_1 = 0.12$	$i_2 = 0.22$	Global	Absolute Stepwise	Global	Absolute Stepwise
Steady state	case 1	case 5	0.97	0.97	0.87	0.87
Zig-zag 30°	case 2	case 6	0.92	1.01	0.89	0.89
Zig-zag 60°	case 3	case 7	1.02	1.00	1.12	1.00
Reversing	case 4	case 8	0.96	1.02	0.74	0.73

Table 3-4 presents the ratio of transverse, R_{A_y} , for each of the four and two calculation methods. For the zigzag 60° flow pattern, A_y is independent of the hydraulic gradient magnitude ($R_{A_y} \approx 1$) for both laboratory and numerical results. The other flow patterns, on the other hand, produced discrepancies between the laboratory results and the numerical simulations. Data from laboratory experiments suggest that A_y was smaller in scenarios of higher hydraulic gradient (i_2) than for the lower hydraulic gradient (i_1), with R_{A_y} ranging from 0.6 to 0.8; conversely, numerical data suggests that A_y increases with increasing gradient magnitude (R_{A_y} varies from about 1.2 in the zigzag 30° flow regime, to more than 2.6 for the steady state and reversing scenarios). Note that R_{A_y} ratios that deviate the most from a value of one also coincide with scenarios where A_y is smallest (steady state and reversing flow), because the transverse component of flow was also small (see Figure 3-7 and Figure 3-10).

Table 3-4. Comparison of the transverse macrodispersivity A_y between high and low hydraulic gradients ($i_2 = 0.22$ and $i_1 = 0.12$, respectively). The ratio R_{A_y} is shown for each pair of scenarios where the configuration of the flow direction was the same (e.g. case 5 vs. case 1, case 6 vs. case 2, and so on). A value of one indicates identical results for both gradients.

Flow scenario	Gradient magnitude, i		$R_{A_y} = A_{y_2} / A_{y_1}$			
			Laboratory experiments		Numerical simulations	
	$i_1 = 0.12$	$i_2 = 0.22$	Global	Absolute Stepwise	Global	Absolute Stepwise
Steady state	case 1	case 5	0.59	0.59	2.55	2.55
Zig-zag 30°	case 2	case 6	0.84	0.70	1.21	1.17
Zig-zag 60°	case 3	case 7	0.97	0.98	0.94	0.96
Reversing	case 4	case 8	0.66	0.70	2.41	2.69

2.4 Effect of the flow pattern

The effect of the four flow scenarios (steady state, zigzag 30°, zigzag 60°, and reversing) on the magnitude of the velocity vector ($||\mathbf{v}||$) was shown in Figure 3-9 for the experimental results, and in Figure 3-12 for the numerical results. As shown in Figure 3-9

and in Figure 3-12, velocity estimated from the *Absolute Stepwise* method is identical for all flow scenarios for a given gradient magnitude i . This indicates that the groundwater velocity is constant for any given time step when direction is ignored. This is a necessary result of the experimental setup: the gradient is maintained constant at all times even if the direction changes. Conversely, when the *Global* method is used for transient flow scenarios where the mean flow direction is rotated, $\|\mathbf{v}\|$ is smaller than for the steady case, proportionally to the cosine of the rotation angle θ . For example, $\|\mathbf{v}\| = 1.54 \text{ cm min}^{-1}$ for the steady state case in Figure 3-9, but the zigzag case with $\theta = 30^\circ$ shows $\|\mathbf{v}\| = 1.37 \text{ cm min}^{-1}$, which corresponds almost exactly to the predicted 1.33 cm min^{-1} ($1.54 \cos(30^\circ)$). Similarly for the case when $\theta = 60^\circ$, $\|\mathbf{v}\| = 0.80 \text{ cm min}^{-1}$ compared to the predicted 0.77 cm min^{-1} ($1.54 \cos(60^\circ)$). For the reversing case, $\|\mathbf{v}\| = 0.50 \text{ cm min}^{-1}$, which is exactly one third of that for the steady case; this is due to the relative duration of the sub-intervals, the forward flow periods lasting twice as long as the reverse flow intervals. This illustrates that the *Global* calculation method is “macroscopic” in the sense that it integrates out the short-lived variations in the flow field. The greater the deviations from the steady state velocity, the lesser the apparent “macrovelocity”.

Figure 3-7 and Figure 3-10 illustrate that changes in the hydraulic gradient direction have a significant impact on macrodispersivity. In general, lateral changes in the flow direction (i.e. “zigzag” cases) result in a decline in the component of macrodispersivity longitudinal to the apparent “macrovelocity” (A_x), while A_y increases as the transverse components of flow “macrovelocity” become increasingly large. This behaviour is apparent in both experimental and numerical results, irrespective of the calculation method; although the *Global* method tends to damp this effect. Similarly, periodic reversals in the hydraulic gradient (“reversing” cases) have a large influence on macrodispersivity estimates. When using the *Global* calculation method, the apparent macrodispersivity was much larger than the steady state case, both longitudinally and transversely; whereas the reverse was true when using the *Absolute Stepwise* method. Moreover, the effect of directional flow transients on A_y is much more pronounced in the numerical simulations than for laboratory experiments (this will be elaborated upon in the last section in this chapter).

2.5 Effect of the calculation method

The method selected to estimate $\|\mathbf{v}\|$ and D_{ij} – and consequently A_{ij} – has a significant impact on the results when the solute plume is subjected to flow transients. When the plume experiences variations in the hydraulic gradient direction, at least one of the components of the plume velocity ($\bar{v}_x$ or $\bar{v}_y$) may temporarily become negative. The *Global* method, because it averages out negative and positive velocities, underestimates the true velocity experienced by the plume and does not completely capture the evolution over time of the plume centroid. As a result, macrodispersivity estimates obtained using the *Global* method are overestimated compared to results derived from the *Absolute Stepwise* method.

When the direction of the hydraulic gradient is not steady, the *Global* method underestimates $\|\mathbf{v}\|$ compared to the *Absolute Stepwise* method because only the net plume movement is taken into account. While the *Global* method is the most commonly used method, it is increasingly inaccurate as flow transients affecting the flow regime become more significant. This can be seen from both laboratory and numerical results (see Figure 3-9 and Figure 3-12, respectively). This trend becomes more apparent as the intensity of the flow transience increases. For instance, $\|\mathbf{v}\|$ derived from the *Global* method for the steady, zigzag 30°, zigzag 60° and reversing scenarios is equal to 1.54, 1.37, 0.80 and 0.50, respectively. Consider for example the hypothetical case where the hydraulic gradient is periodically reversed, but where the periods of flow reversals are on average as long as periods of forward flow, so that the plume centroid returns to its original location. The net plume displacement would therefore be near zero, and would result in a *Global* estimate of $\|\mathbf{v}\|$ also close to zero. Plume spreading due to hydrodynamic dispersion obviously continues to increase regardless of whether the hydraulic gradient is along the positive or negative direction (+x or -x, respectively). Consequently, macrodispersivity estimated from the *Global* method using (3.2) would be infinitely large. This is clearly not representative of the migration history of the solute plume. When a dissolved solute is subjected to significant variations in the hydraulic direction, the *Absolute Stepwise* method therefore seems more appropriate.

A comparison of the macrodispersivity estimated using the *Global* vs. *Absolute Stepwise* methods clearly shows that the former approach leads to an overestimation when the mean flow is unsteady. This is illustrated for laboratory results by comparing Figure 3-7(a) vs. Figure 3-7(b), and for numerical results in Figure 3-10(a) vs. Figure 3-10(b). Despite the significant differences in macrodispersivities estimated from the two methods, the ratio A_y / A_x for a given scenario is almost identical, regardless of the calculation method used. Note that this is true for laboratory as well as numerical results, although the ratio varies considerably between the two datasets (see §2.6).

The uncertainty associated with each of the parameters discussed above is plotted as error bars in the above figures. These are generally much smaller when quantities are calculated using the *Global* method instead of the *Absolute Stepwise* method. Moreover, this trend becomes increasingly true as the flow transients become more significant. This can be explained by the fact that the latter calculation method incorporates the small-scale variability induced by the variations in the hydraulic gradient, whereas the *Global* method does not.

2.6 Laboratory vs. numerical results

One of the main objectives of this study is to assess the ability of mathematical models to accurately reproduce the physical conditions observed in the laboratory experiments. This is done in this section by comparing results derived from numerical simulations with the laboratory results.

The magnitude of the velocity vector $\|\mathbf{v}\|$ estimated from laboratory or numerical results is almost identical for a given scenario (see Figure 3-9 and Figure 3-12, respectively). Note, however, the slight overestimation in the numerical simulations of the plume velocity calculated using the *Absolute Stepwise* method. This is likely the result of small inaccuracies in the boundary conditions specified as input to the numerical model, or in measuring the gradient in the laboratory.

The effect of flow transience on transverse plume dispersion is significantly more pronounced in numerical results than for laboratory experiments. As was discussed

previously (see §2.3), gradient magnitude has little impact on A_x and A_y . The similarities and differences between laboratory and numerical results follow the same trends within a specified group of scenarios, where the hydraulic gradient is constant. The following discussion will therefore only look at the “low gradient” scenarios, with $i = 0.1211$ (cases 1-4).

For the steady state scenario, estimates of A_x and A_y from laboratory and numerical results coincide (with $A_x = 0.048$ cm and $A_y = 0.002$ - 0.003 cm for both). This is not surprising considering that the simulations were calibrated with the steady state case. When flow is rotated (zigzag 30° and 60° scenarios), on the other hand, A_x decreases while A_y increases relative to the steady state scenarios. The decrease in A_x is slightly more pronounced in laboratory experiments compared to numerical simulations, but the increase in A_y is much more pronounced in the numerical results than in the laboratory data. This trend is visible from results obtained from both the *Global* and *Absolute Stepwise* calculation methods.

The *Global* estimates of A_x and A_y are both drastically enhanced by reversing flow conditions, and this behaviour is much more pronounced in numerical results. For example, A_x increases between the steady and reversing cases by a factor of 1.8 in laboratory results, compared to a factor of 2.9 for numerical results (see Figure 3-7(a) and Figure 3-10(a), respectively). Similarly, A_y is enhanced by reversing flow conditions by a factor of 2.6 to 3.9. As discussed previously, however, the effects of flow transience on plume spreading are artificially enhanced by the *Global* calculation method because this approach underestimates plume velocity by averaging out the net plume displacement rather than accounting for its absolute movement. Consequently, the *Absolute Stepwise* results are expected to provide a more accurate description of the plume behaviour, for results obtained from both laboratory experiments and numerical simulations.

Comparing the laboratory vs. numerical results obtained using the *Absolute Stepwise* method (Figure 3-7(b) vs. Figure 3-10(b)) provides a more accurate representation of the “true” behaviour of the dissolved solute because it explicitly includes the effects of all variations in the flow field. As in the *Global* estimates, the decrease in A_x due to a rotating hydraulic gradient is more important in laboratory results than in numerical

simulations, while the opposite is true for A_y (it increases with increasing rotation angle, and this trend is more pronounced in the numerical results). In numerical results, the reduction in A_x for the zigzag 30° is in line with the rotation angle ($A_x = 0.0412 \approx 0.0479 \cos(30^\circ) = 0.0413$); and similarly for the zigzag 60° case with $A_x = 0.0207 \approx 0.0479 \cos(60^\circ) = 0.0239$ cm. A_x estimated from laboratory results (Figure 3-7 (b)) is approximately 63% smaller under reversing flow conditions than for the steady state case (0.0303 vs. 0.0479 cm, respectively). Conversely, A_x estimated from the numerical results (Figure 3-10(b)) indicates that reversing flow has no influence on A_x (0.0465 cm).

These results indicate a sizeable discrepancy between the numerical simulations and laboratory results for the transient cases. While no clear reason for this discrepancy could be identified, it suggests that there may be a “memory effect” present in the laboratory experiment that is not present in the simulations: the numerical model treats each new time step as though the solute plume sees a fresh, new portion of the porous medium that is only partially correlated to that of the previous time step. This would be the case irrespective of the flow transience. The reality of the laboratory may be somewhat different, in particular when flow direction is reversed. When the real plume travels back in the exact reverse direction, it does not see a fresh realization of the porous medium, but rather a realization that is more highly correlated with that of the previous time step. The randomness of the mixing process is therefore reduced, which is reflected in the apparent macrodispersivity.

CHAPTER 4 – Experimental and Numerical Results for Heterogeneous Porous Media

“ When the number of factors coming into play in a phenomenological complex is too large, scientific method in most cases fails. One need only think of the weather, in which case the prediction even for a few days ahead is impossible. Nevertheless, no one doubts that we are confronted with a causal connection whose causal components are in the main known to us. Occurrences in this domain are beyond the reach of exact prediction because of the variety of factors in operation, not because of any lack of order in nature.”

“Things should be made as simple as possible, but not any simpler.”

– Albert Einstein

The previous chapter investigated the macroscopic effects of flow transients on hydrodynamic dispersion in a homogeneously permeable porous material. While this provided useful insights into the behaviour of a dissolved tracer in a flow field subjected to temporal transience, the next logical step was also undertaken: to extend this analysis to a more complex, heterogeneous geologic material.

This chapter presents, discusses and interprets the experimental and numerical results for a heterogeneous material that was artificially generated with a prescribed spatial correlation structure. The methodology used to produce this field was presented in Chapter 2. First, a few image sequences and contour plots taken from selected runs are shown to illustrate the very different dispersive behaviour of a heterogeneous medium. Spatial moments and derived quantities, presented in the form of time series, are then discussed in detail for each of the flow scenarios (cases). Macrodispersivity, calculated from the temporal evolution of

the normalized first- and second-order moments, is then characterized for each scenario. Finally, the effects of the flow pattern on macrodispersivity estimated using one of two methods are discussed; this is followed by a comparison between the laboratory and numerical results. Experimental results from the laboratory experiments that were conducted using the heterogeneous porous medium discussed above, along with the corresponding numerical simulations, are presented and analyzed below. First, an overview of the experimental evidence is provided in the following section, in the form of (1) normalized image sequences from laboratory experiments and (2) contoured solute concentrations and equipotentials from numerical simulations. Second, the temporal evolution of the spatial moments up to the fourth order are shown in § 2. These are used to describe the physical characteristics of the migrating solute plumes as they develop during laboratory experiments or numerical simulations. Finally, macrodispersivity estimates calculated using the *Global* and *Absolute Stepwise* methods are presented and discussed in § 3.

1 EXAMPLES OF EXPERIMENTAL RESULTS

1.1 Laboratory experiments

For each of the laboratory experiments, a series of high-resolution greyscale images was acquired at regular intervals $\Delta t = 0.5$ min. Each image (or “slice” in image processing terminology) within such a sequence (“stack”) was then processed to remove defects such as noise and spatial and/or temporal variations in illumination, to produce “normalized” image sequences. Details of the image processing are provided in Appendix B. Normalized pixel values were ultimately converted into solute concentration units and used to calculate spatial moments, which were in turn used to obtain macrodispersivity estimates. These results are shown and discussed in later section of this chapter.

Selected images were extracted from the extensive dataset to provide a visual overview of the temporal evolution of the dissolved solute plumes. These normalized images are shown for cases 9 through 12 in Figure 4-1 to Figure 4-4, respectively.

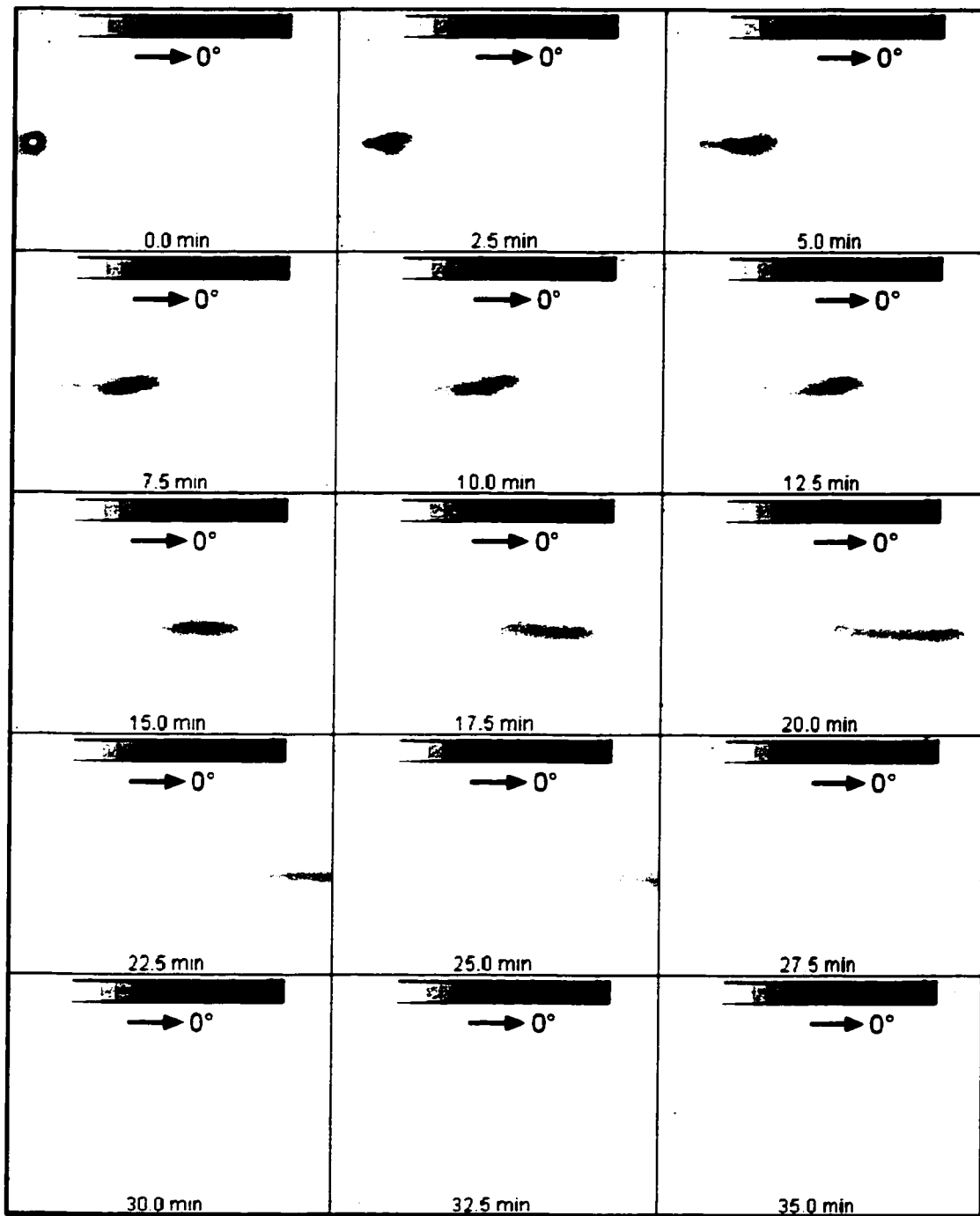


Figure 4-1. Selected images from a typical steady state scenario (case 9). Flow direction is shown by the arrows (the angle is expressed relative to the average longitudinal direction). See Figure 4-6 for plots derived from this sequence.

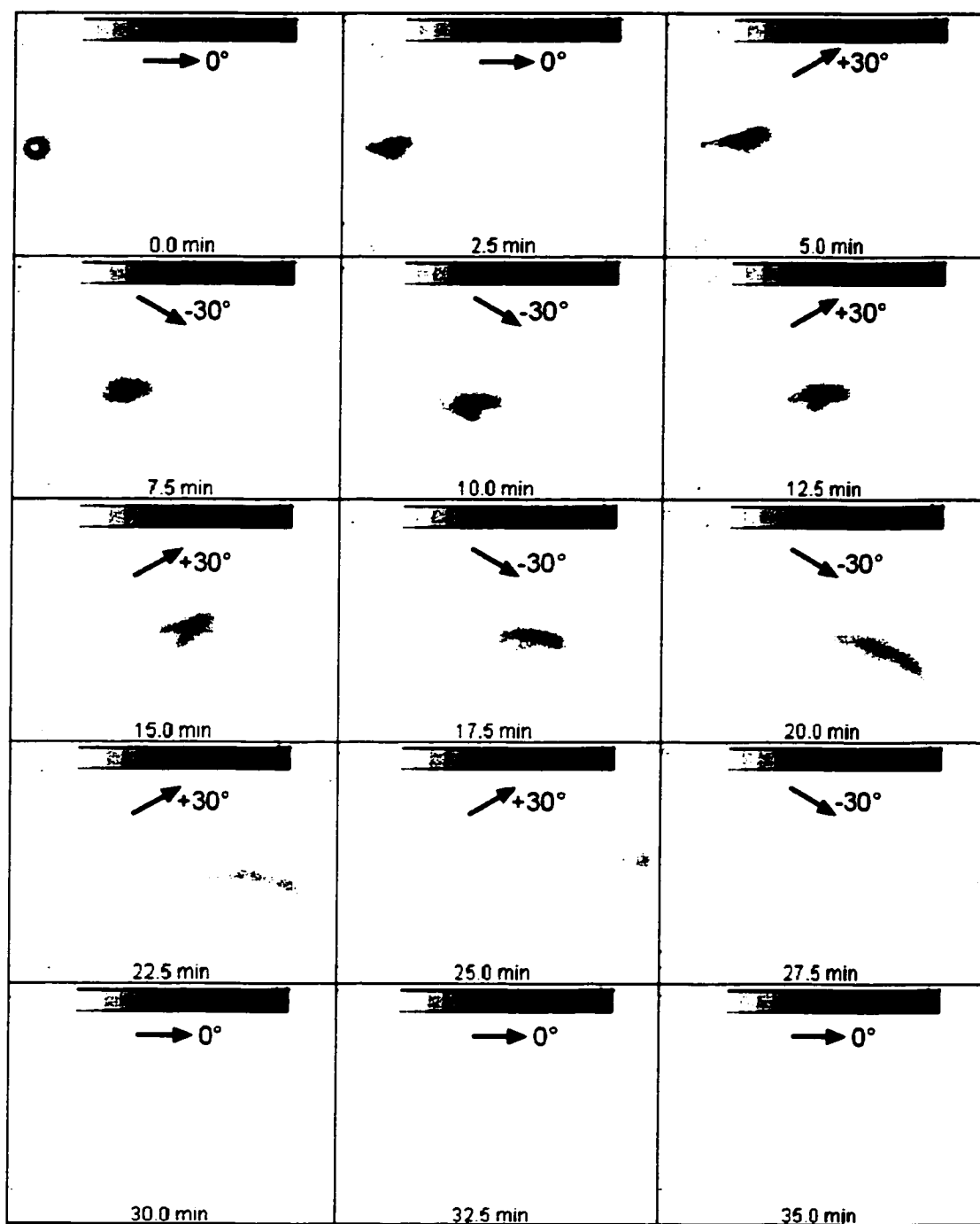


Figure 4-2. Selected images from a typical "zigzag 30°" flow scenario where the hydraulic gradient direction was varied in time at an angle of $\pm 30^\circ$ (case 10). Flow direction is shown by the arrows (the angle is expressed relative to the average longitudinal direction). See Figure 4-7 for plots derived from this sequence.

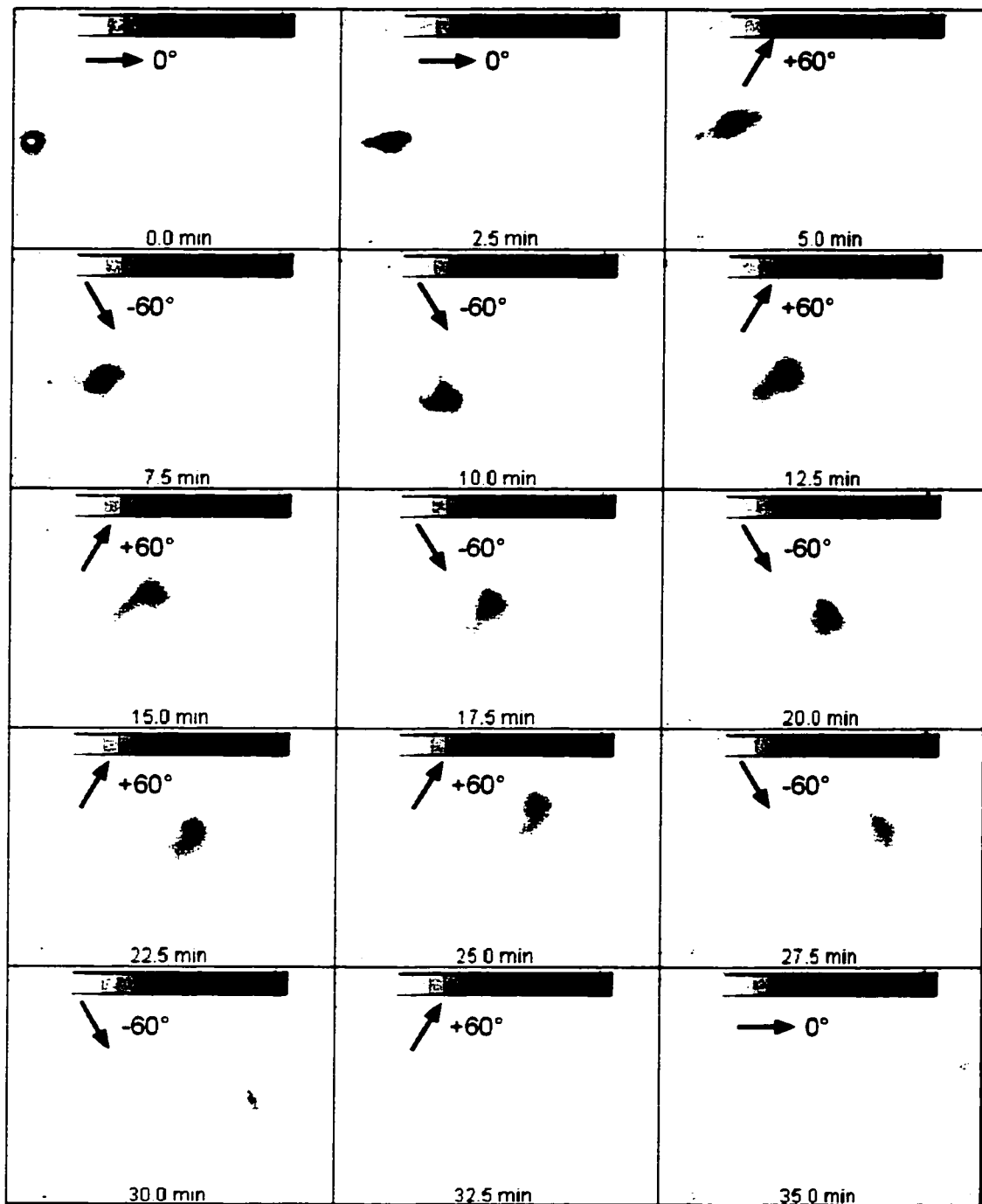


Figure 4-3. Selected images from a typical "zigzag 60°" flow scenario where the hydraulic gradient direction was varied in time at an angle of $\pm 60^\circ$ (case 11). Flow direction is shown by the arrows (the angle is expressed relative to the average longitudinal direction). See Figure 4-8 for plots derived from this sequence.

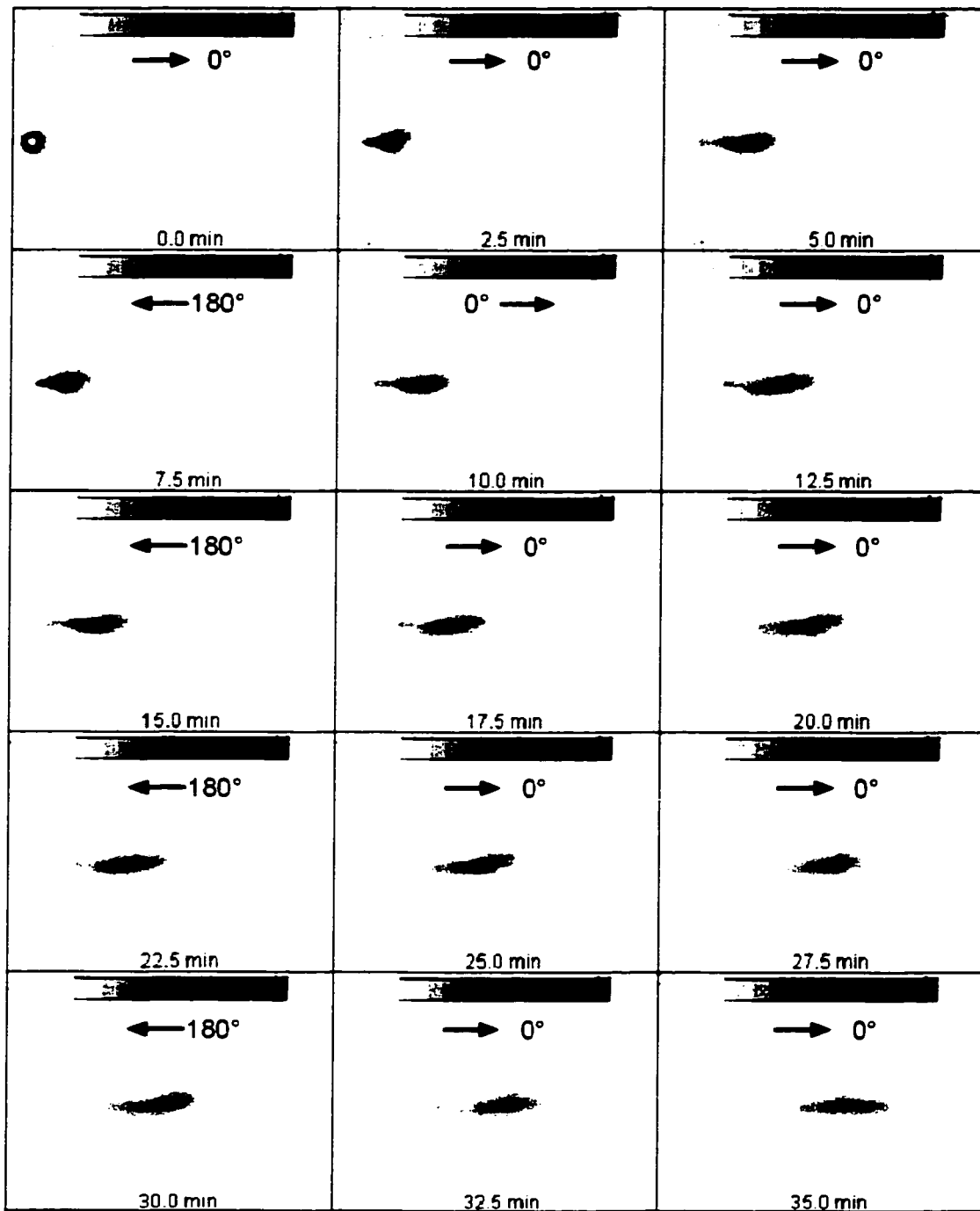


Figure 4-4. Selected images from a typical “reversing” flow scenario where the hydraulic gradient direction was reversed periodically (case 12). Periods of forward flow and reversed flow (parallel to the longitudinal direction, x) were $\Delta t_1 = 10$ min and $\Delta t_2 = 5$ min, respectively. Flow direction is shown by the arrows (the angle is expressed relative to the average longitudinal direction). See Figure 4-9 for plots derived from this sequence.

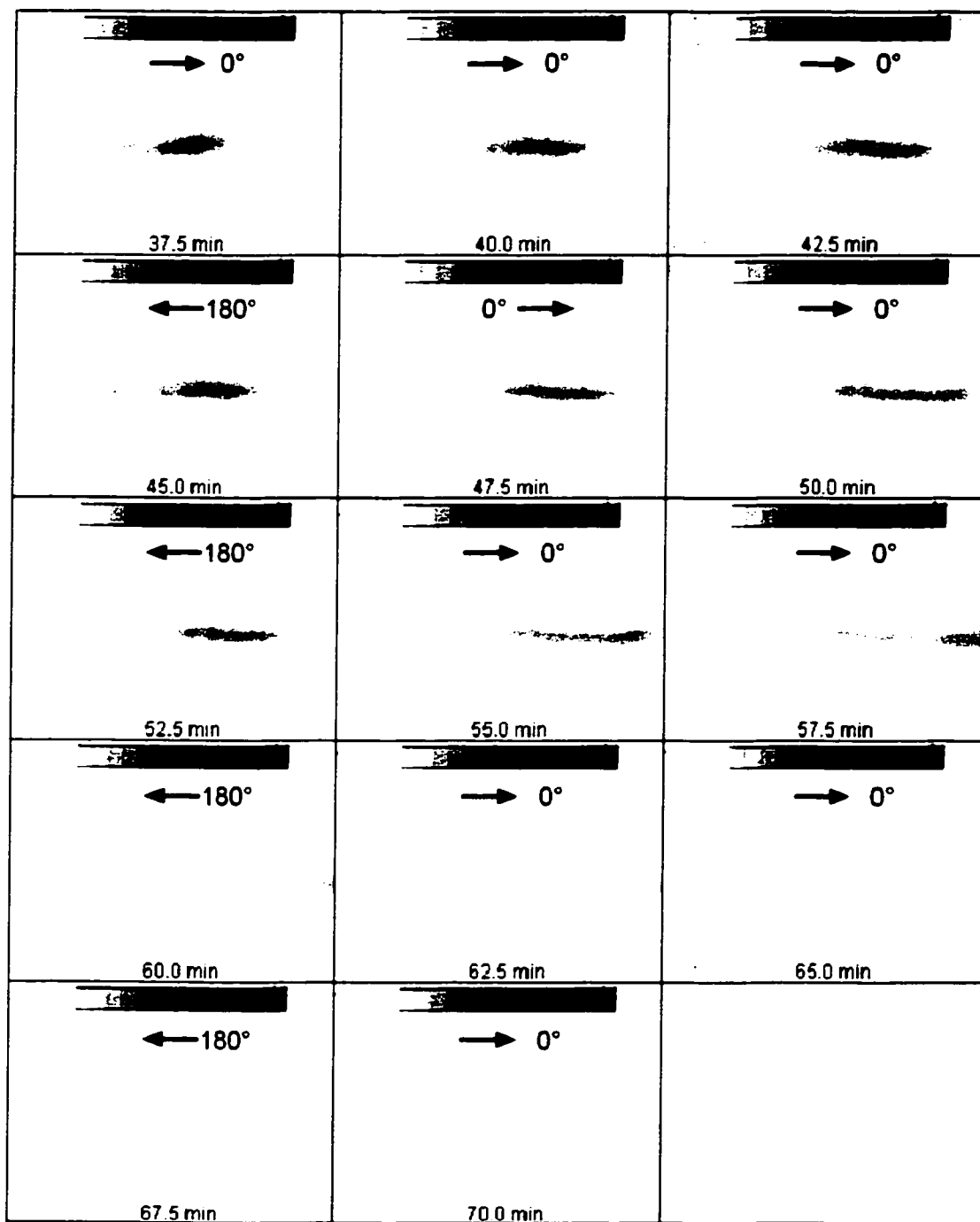


Figure 4-4. (continued)

Selected images from a typical "reversing" flow scenario where the hydraulic gradient direction was reversed periodically (case 12). Periods of forward flow and reversed flow (parallel to the longitudinal direction, x) were $\Delta t_1 = 10$ min and $\Delta t_2 = 5$ min, respectively. See Chapter 2 for details of the transient flow schedule. Flow direction is shown by the arrows (angle expressed relative to the longitudinal direction). See Figure 4-9 for plots derived from this sequence.

1.2 Numerical simulations

Numerical simulations were performed using the same heterogeneous hydraulic conductivity distribution as in the laboratory model. Hydraulic gradients were imposed on the system by specifying constant head boundary conditions at nodes corresponding to the inlet/outlet ports of the flow cell. These constant heads either remained unchanged throughout the duration of the experiment (in the steady state simulation, case 9), or varied in time as required by the transient schedules for cases 10 (zigzag 30°), 11 (zigzag 60°) and 12 (reversing). Refer to Chapter 2 for the model details.

Figure 4-5 shows a typical example of the results obtained from numerical simulations. Steady state equipotentials from case 9 are shown along with the contoured solute distribution 10 minutes after the tracer was released at the injection port location (black circle in the central left portion of the flow cell). Figure 4-5 also includes the nodal velocity vectors for completeness. The initial solute distribution $(C/C_0)_i$ near the injection port was the same as that used in the previous chapter, expressed as unitless concentration. These were later translated into actual concentrations in the moment-calculating algorithm, by multiplying the relative simulated concentration C/C_0 at each node by the known injection concentration $C_0 = 15.47 \text{ mg L}^{-1}$. As a result of this linear conversion, the spatial moments reported in the following section were calculated using “true” nodal concentrations expressed in units of mg L^{-1} . The choice of C or C/C_0 is irrelevant to the moment calculations, since all moments are normalized with the zero-order moment (the total mass in the system).

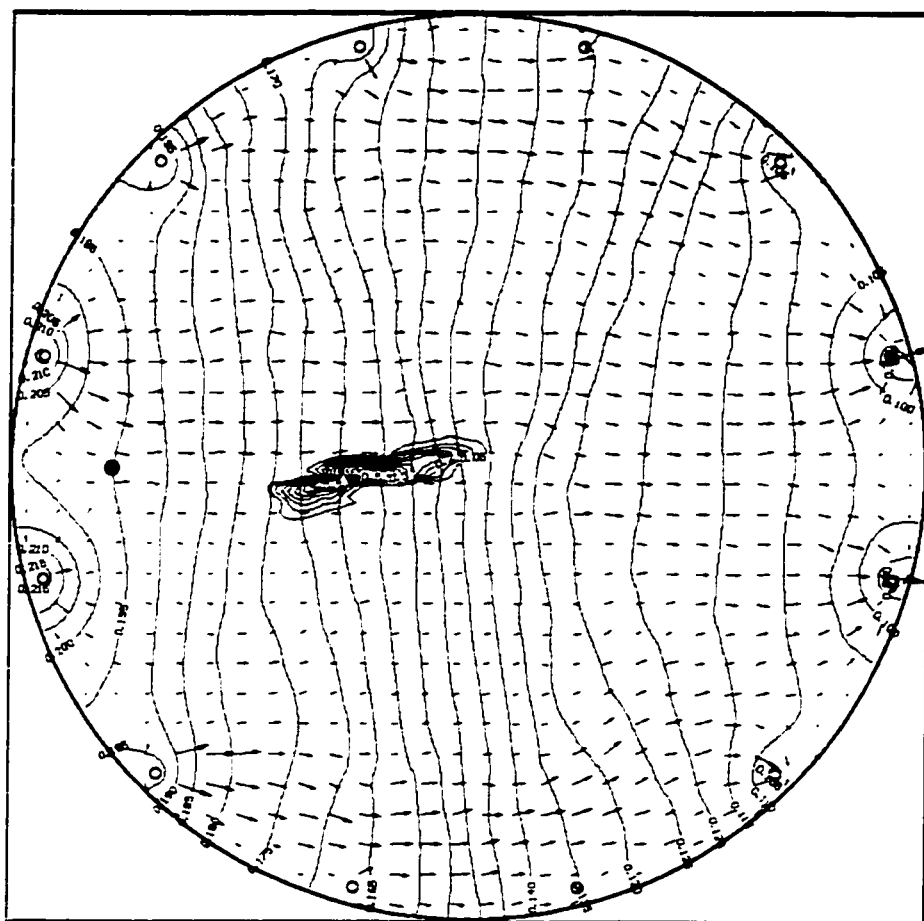


Figure 4-5. Hydraulic head and solute distributions from the steady state numerical simulation in the heterogeneous porous medium (case 9) at $t = 10$ min. Steady state equipotentials are displayed using a contour interval of 0.005 m, and range from $h = 0.217$ at the upstream (leftmost) inlet/outlet ports to $h = 0.089$ m at the downstream (rightmost) ports. Flow direction is generally from left to right, as indicated by the velocity vectors (arrows), whose length is proportional to the groundwater velocity at their origin. Relative solute concentration contours (C/C_0) are expressed using a contouring interval of 0.05. The maximum relative concentration is approximately 0.63 ($C_{max} = 9.7 \text{ mg L}^{-1}$). The solid black circle on the left corresponds to the location of the injection port.

2 SPATIAL MOMENTS AND DERIVED QUANTITIES

Spatial moments up to the fourth order were calculated at 0.5 minute intervals from dissolved solute concentration distributions obtained during the laboratory experiments and numerical simulations. Physically meaningful quantities (e.g. total mass, centroid location, spatial variance, etc.) were derived from these spatial moments, and are plotted over the next eight pages for cases 9 through 12 in Figure 4-6 to Figure 4-9, respectively. Each figure

includes plots against time of (a) total mass, (b) centroid location, (c) spatial variance, (d) plume orientation and elongation, (e) coefficient of skewness, and (f) kurtosis. To facilitate comparisons between laboratory and numerical results, the time series are plotted side by side. Details on spatial moment calculations and their significance in the context of this study are provided in Appendix C. The time behaviour of the spatial moments and associated quantities are discussed in detail in the sections following the graphs.

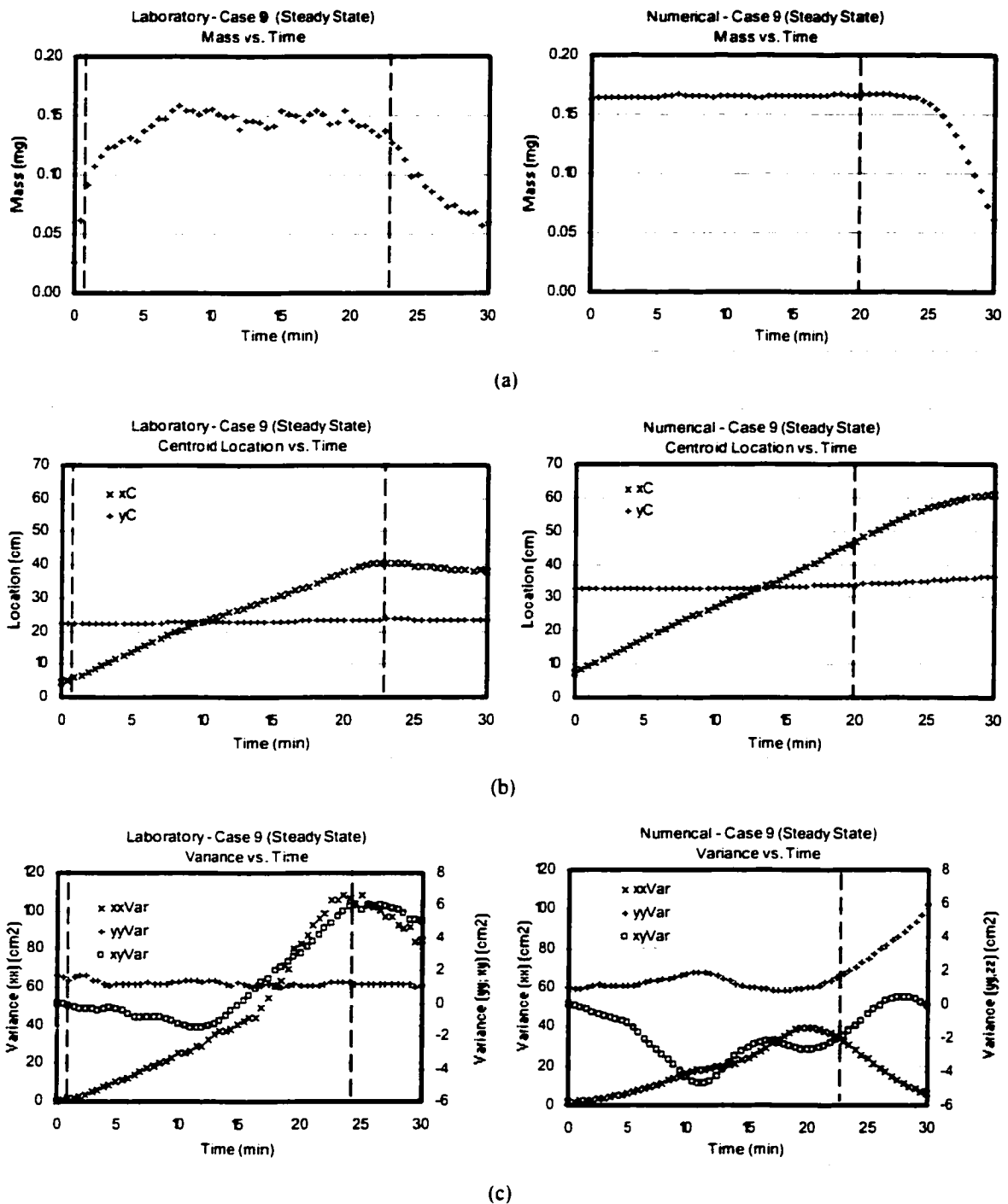
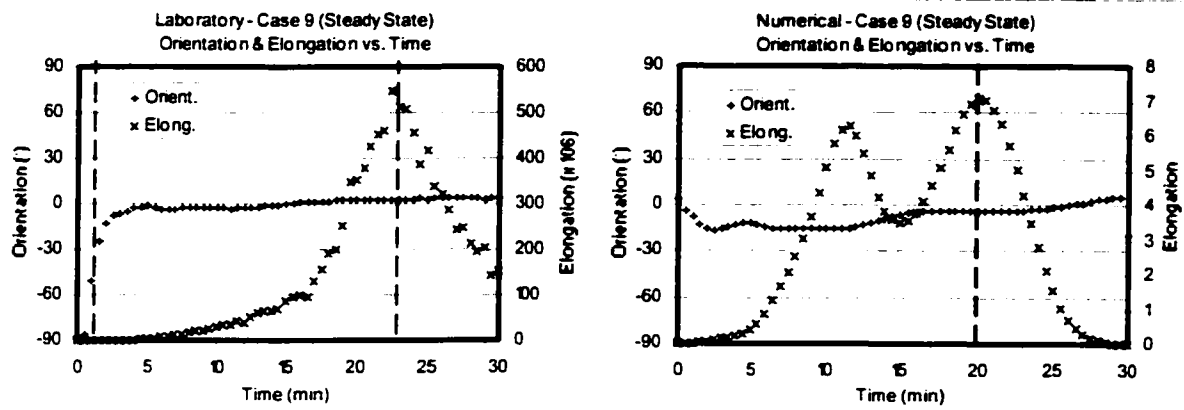
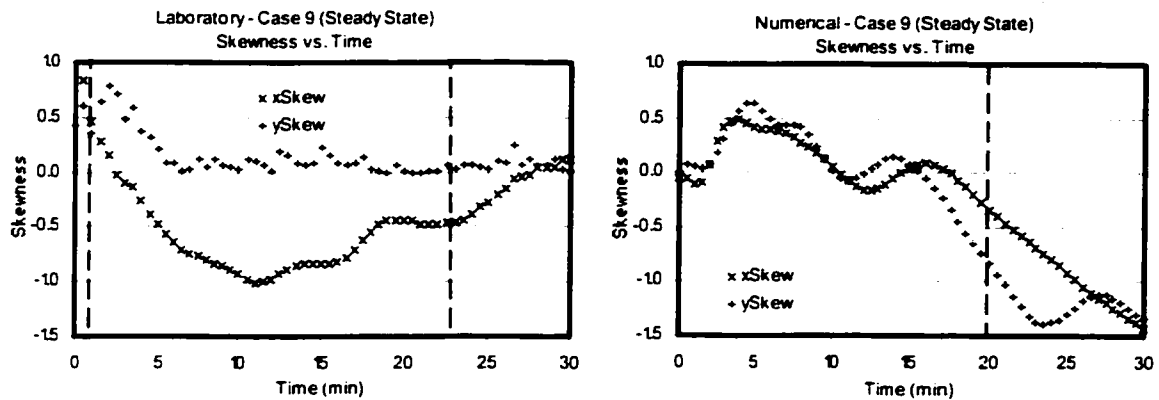


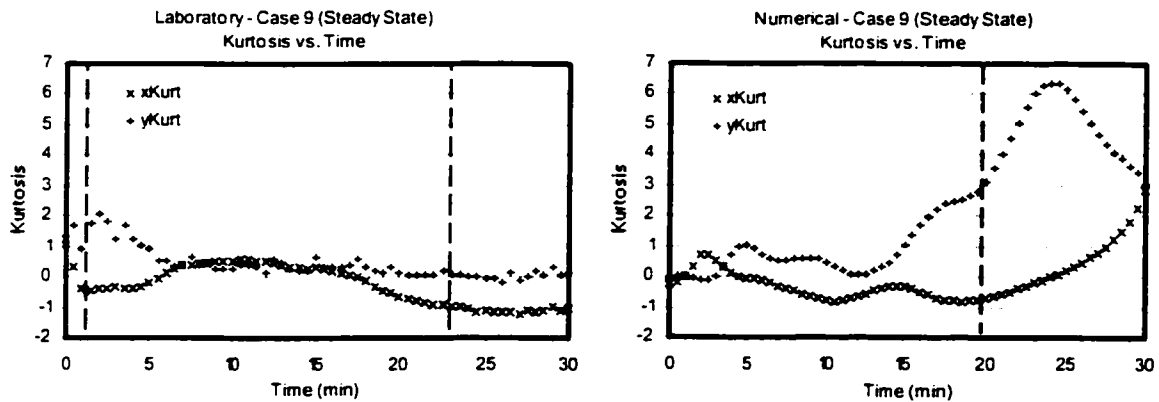
Figure 4-6. Results from case 9 (steady state). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. See Figure 4-1 for sample images from this scenario.



(d)



(e)



(f)

Figure 4-6. (continued)

Results from case 9 (steady state). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the vertical dashed lines. See Figure 4-1 for sample images from this scenario.

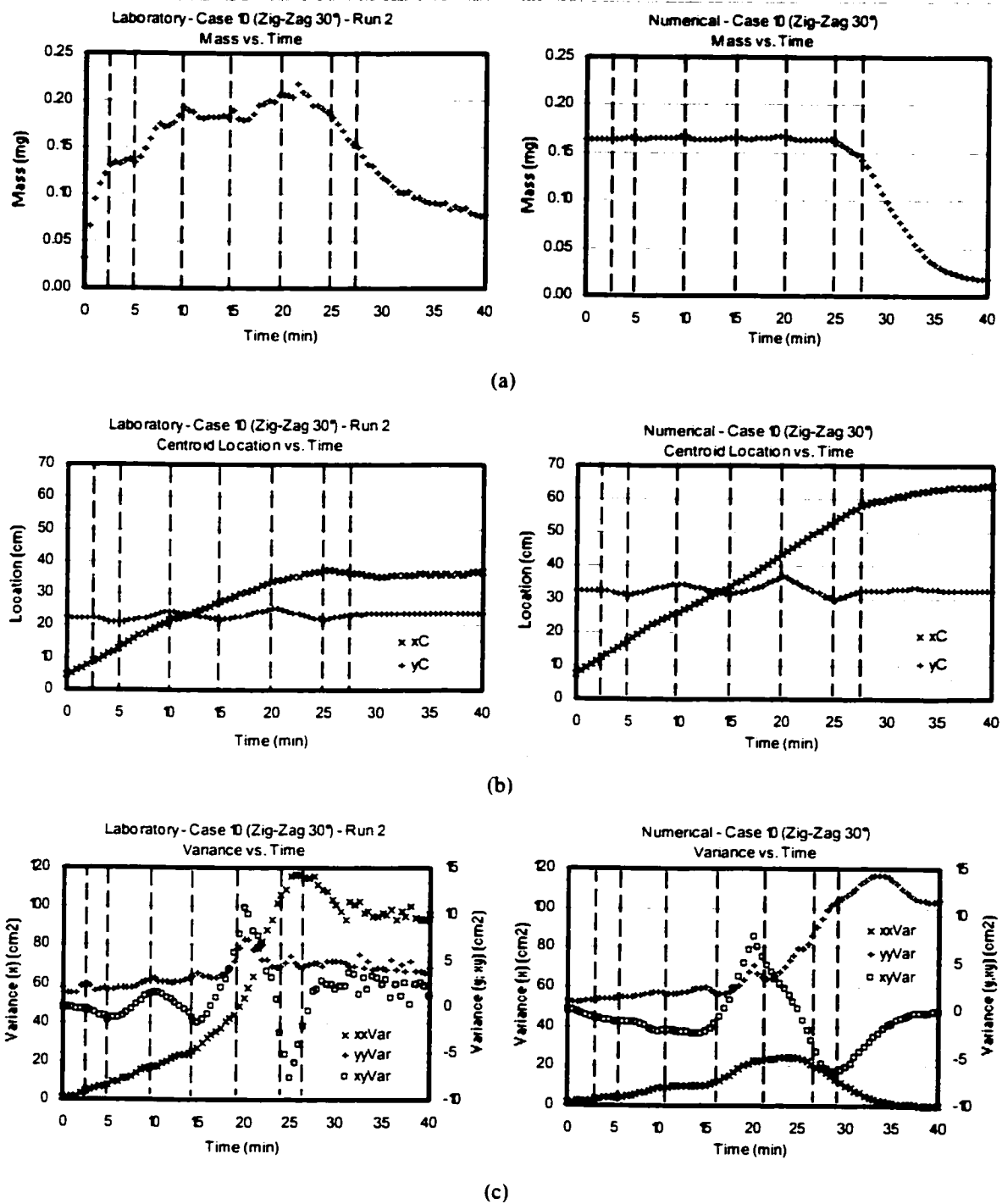


Figure 4-7. Results from case 10 (zigzag 30° scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the first and last vertical dashed lines; others indicate direction changes. See Figure 4-2 for sample images from this scenario.

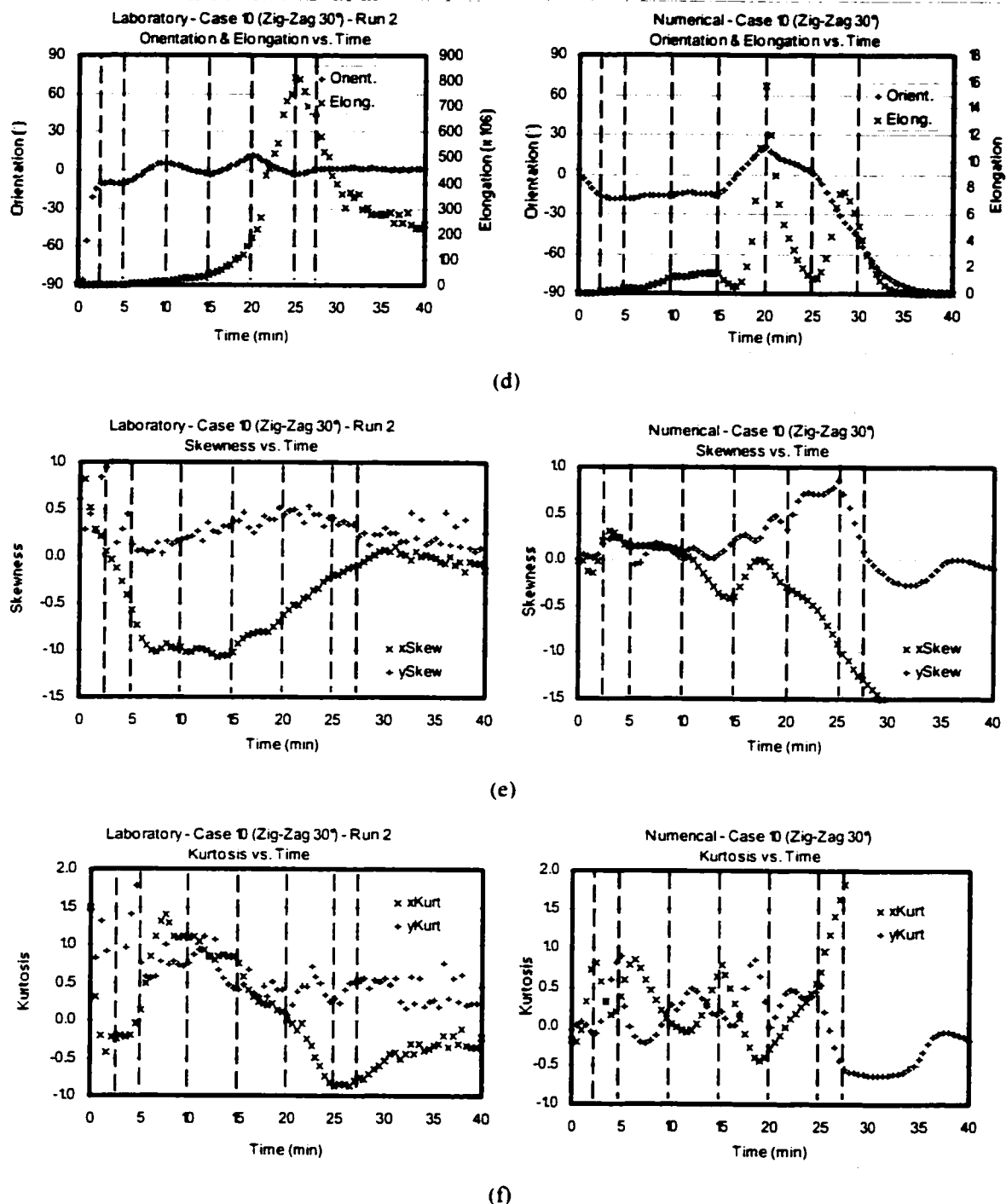


Figure 4-7. (continued)

Results from case 10 (zigzag 30° scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (7) when the plume was in range includes data between the first and last vertical dashed lines; others indicate direction changes. See Figure 4-2 for sample images from this scenario.

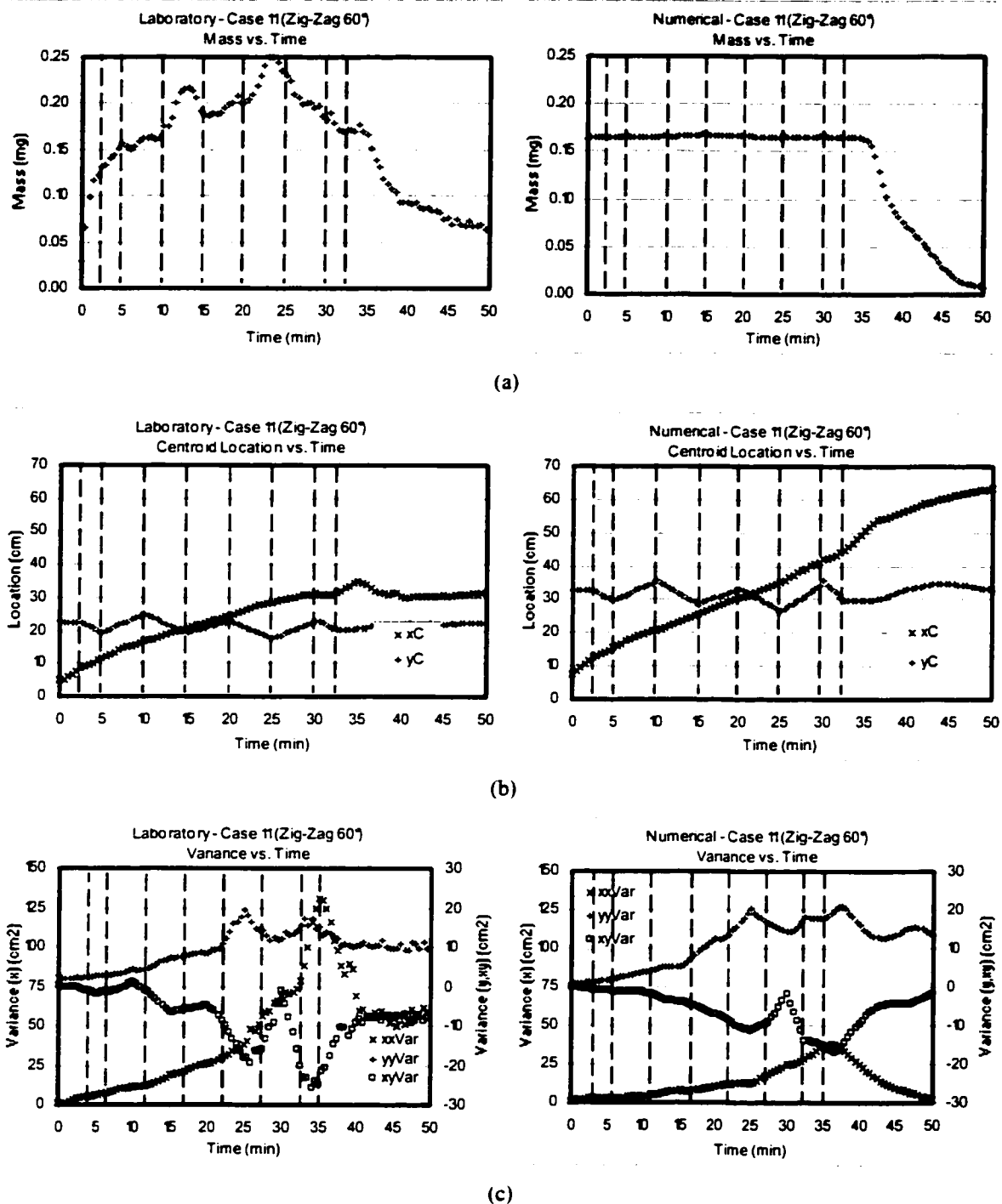


Figure 4-8. Results from case 11 (zigzag 60° scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The “acceptable” time interval (T) when the plume was in range includes data between the first and last vertical dashed lines; others indicate direction changes. See Figure 4-3 for sample images from this scenario.

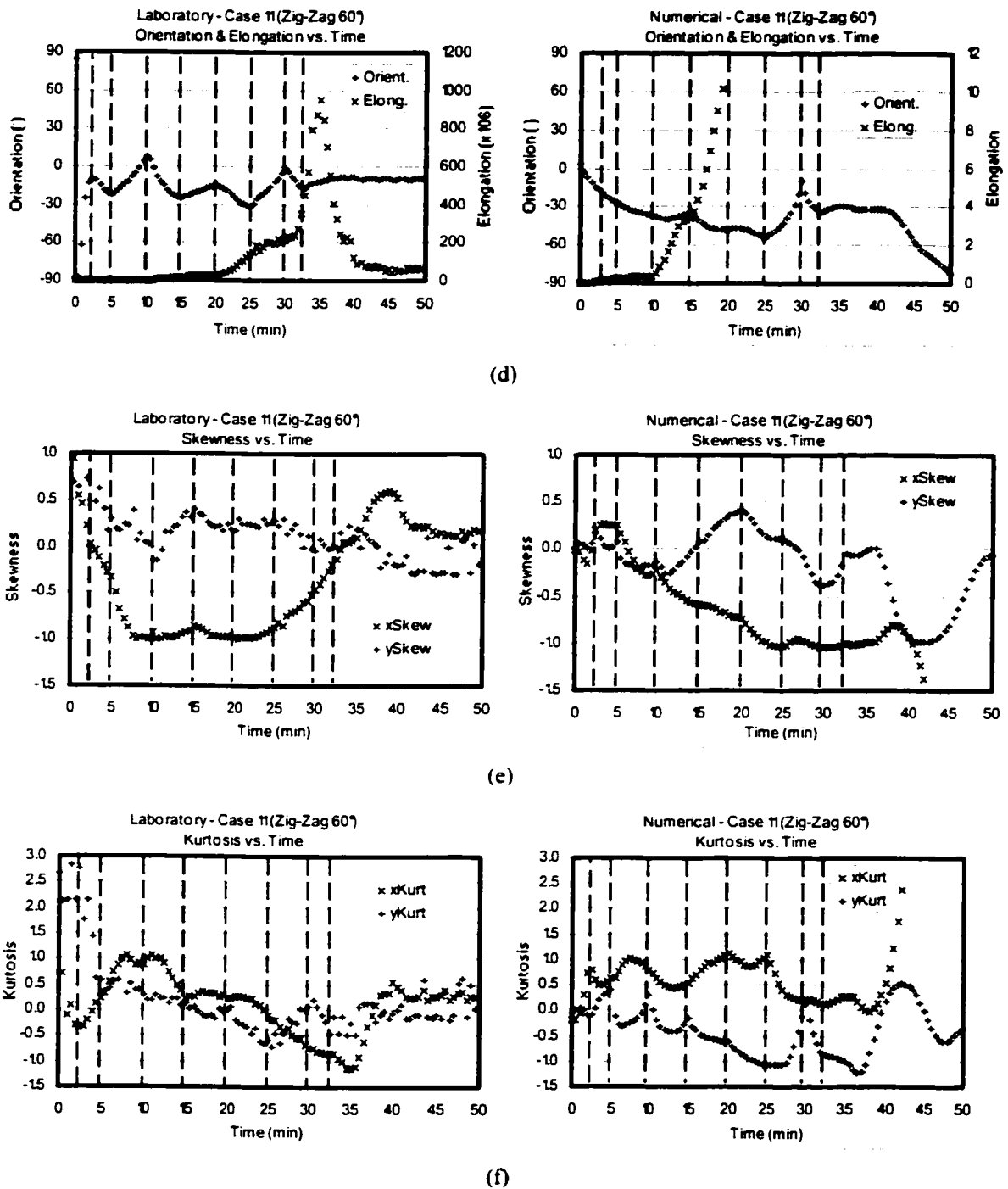


Figure 4-8. (continued)

Results from case 11 (zigzag 60° scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the first and last vertical dashed lines: others indicate direction changes. See Figure 4-3 for sample images from this scenario.

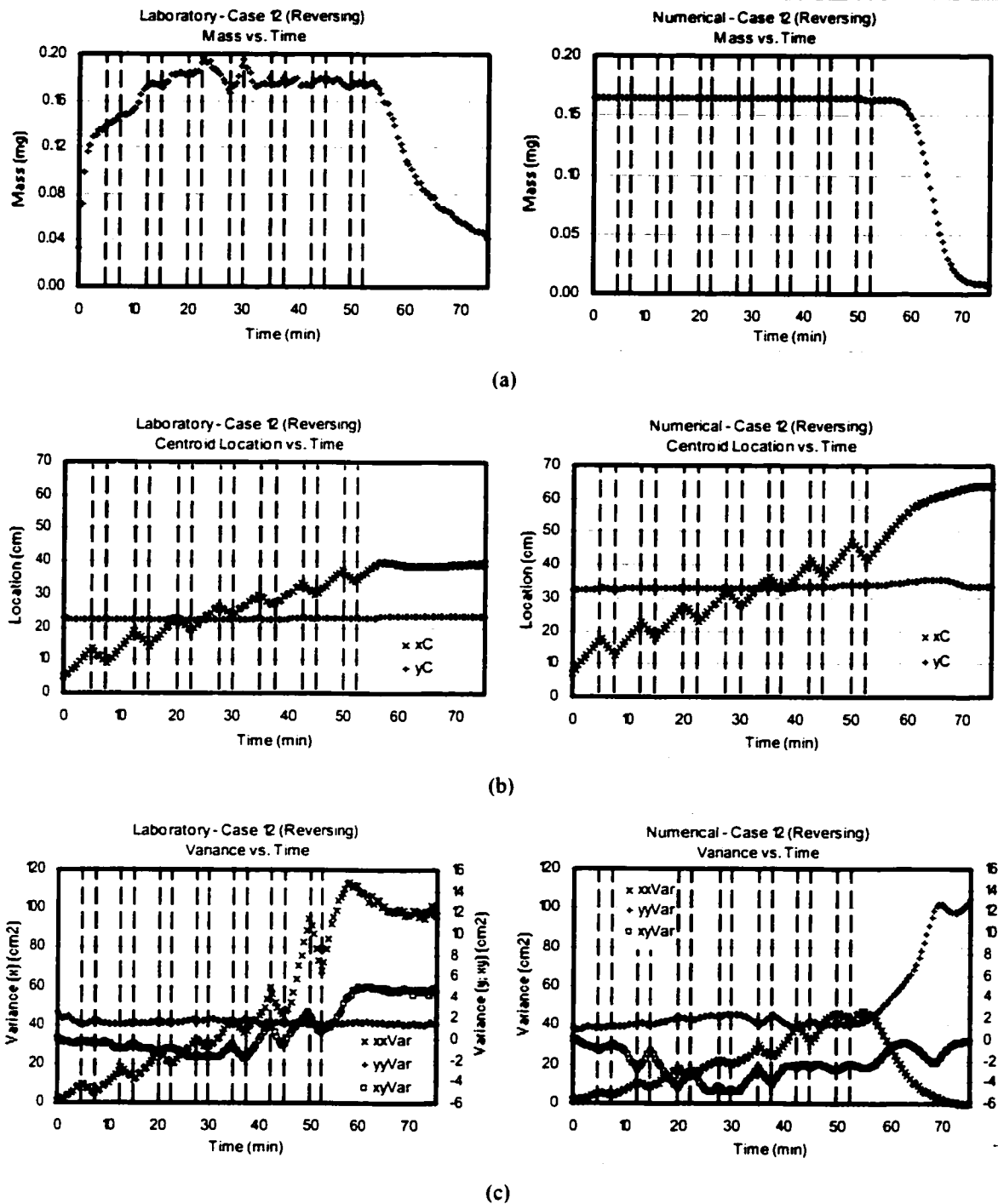


Figure 4-9. Results from case 12 (reversing scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the first and last vertical dashed lines; others indicate direction changes. See Figure 4-4 for sample images from this scenario.

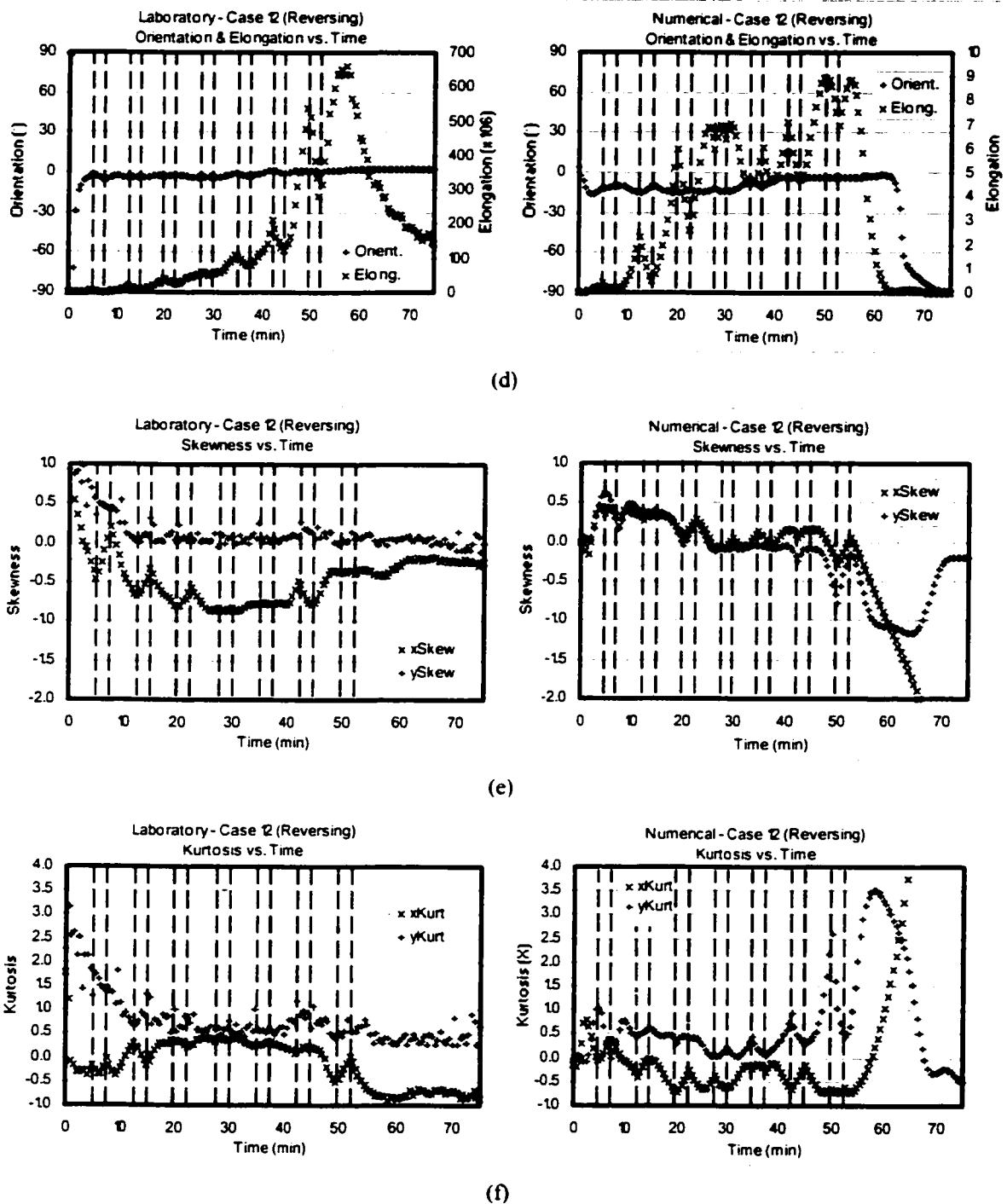


Figure 4-9. (continued)

Results from case 12 (reversing scenario). The time series show parameters derived from spatial moments of orders 1-4 calculated from laboratory (left) and numerical (right) results. These include (a) total mass, (b) location of the centre of mass, (c) spatial variance, (d) orientation, (e) coefficient of skewness, and (f) kurtosis. The "acceptable" time interval (T) when the plume was in range includes data between the first and last vertical dashed lines; others indicate direction changes. See Figure 4-4 for sample images from this scenario.

2.1 Mass balance

Spatial moments of order zero (M_{00}) were integrated over the entire flow domain and used to estimate the total tracer mass present in the system at any given time. Plots of solute mass against time were presented for cases 9 to 12 in part (a) of Figure 4-6 to Figure 4-9, respectively. The mass estimated from laboratory and numerical results was compared with the known injected mass $M = 0.01 \text{ L} \cdot 15.47 \text{ mg L}^{-1} = 0.155 \text{ mg}$.

In the case of the numerical simulations, total mass was essentially constant at 0.165 mg throughout most of the duration, until the bulk of the plume had reached the end of the flow cell. At that point, mass removal via the downstream ports is clearly visible from the sharp decline in total mass. As expected, this estimate obtained from the spatial moments compared favourably with the total mass calculated by the transport code MT3D (0.163 mg). The slight discrepancy (approximately 5%) compared to the known injected mass (0.163/0.155) is likely the result of the way the initial concentration distribution was estimated. Recall from Chapter 2 that the initial conditions for the transport simulation had to be estimated from the experiments conducted in a homogeneous porous medium because of the obstruction in the pictures caused by the injection port. This approximation may have not been completely applicable in the heterogeneous porous medium.

The temporal evolution of the total mass in the laboratory experiments behaved in a more complex fashion. Initially, total mass is below the expected 0.155 mg. In the steady state and reversing scenarios (cases 9 and 12, respectively), mass increases from about 0.12 mg until it reaches a plateau. On the other hand, the solute mass appears to increase continuously for both zigzag scenarios (cases 10 and 11), even reaching a value more than 50% higher than expected (see Figure 4-8(a)).

Three possible causes were identified to account for the apparent extra mass. First, the mass “gain” might be explained by the fact that the zigzagging plumes encountered a greater portion of the porous medium, which includes some lower permeability (darker) zones. Pixel concentrations in those “dark” areas were artificially inflated because of the noise introduced when images were normalized. The source of the “shot noise” is thought to be related to contrast problems, as discussed in Chapter 2. A second possible reason for the

extra mass is due to the method used by the image processing plugin used to calculate moments at each time step. Moments are computed using only those pixels located within a local ROI, an oval whose dimensions are based on the solute distribution itself. As the plume becomes increasingly large in the vertical direction, the ROI also incorporates a greater proportion of the porous medium, and thus incorporates more pixels containing the artificially high concentrations of darker fine-textured materials. The third possible cause for the mass balance errors is linked with the method used to calibrate the images. Normalized pixel values were calibrated using the polynomial function used for the homogeneous experiments of Chapter 3. Recall from Appendix B that this calibration function was obtained empirically by fitting numerically simulated concentrations to experimental data for a simple (homogeneous, steady state) scenario. Normalized pixel values were thus converted from “Gel Optical Density” units into concentration units. It is likely that this function was not entirely suitable to calibrate images from the heterogeneous experiments. Note that due to the complexity of the heterogeneous porous medium, this approach was not replicated and the previous function was used instead.

The late time portion of all laboratory experiments offers interesting observations (graphs on the left). As in the numerical simulations, the solute mass declines rapidly once the bulk of the plume reaches the discharge area. But unlike the numerical simulations, the mass removal rate gradually becomes slower, giving rise to a tail end whose slope gradually becomes less pronounced as the total mass approaches zero, but without ever reaching it. This peculiar behaviour is seen in all scenarios, and appears to result from a small but significant portion of the tracer being trapped and then gradually released by the low permeability zones that the plume travels through. This effect is likely caused by retardation due to a multi-modal velocity distribution, and diffusion-controlled transport in the slow zones. The precise cause could not be determined with certainty from the available experimental evidence. A batch experiment was conducted using size class 3 beads and showed that the glass beads had no affinity for sorption with the tracer (see Appendix A).

2.2 Centre of mass (centroid) and velocity

The temporal evolution of the plume's centre of mass, derived from first-order spatial moments, is plotted in part (b) in Figure 4-6 to Figure 4-9. As expected, centroid evolution is strongly influenced by the nature of the flow pattern governing groundwater flow.

The mean longitudinal position of the plume x_c increases relatively steadily over time for the steady state and both zigzag scenarios (cases 9 to 11); for case 12 (reversing flow pattern), the slope of x_c vs. t alternates between positive and negative values, travelling approximately twice as far during the "forward" periods. The only obvious differences that can be seen when comparing laboratory results to numerical simulations is the fact that plume migration is significantly faster in the simulation results than in the laboratory data.

In the transverse direction (y_c vs. t), a subtle upward plume movement can be observed in the steady state and reversing scenarios (cases 9 and 12, respectively), as a result of the plume preferentially following the zone of higher hydraulic conductivity. The hydraulic conductivity distribution (see Chapter 2) shows a "fast" zone, located slightly above the horizontal location of the injection port, spanning across most of the flow domain in a left to right orientation, parallel to the mean flow direction for all scenarios. For the zigzag scenarios (cases 10 and 11), y_c moves up and down due to the transient variations imposed on the transverse hydraulic gradient.

2.3 Spatial variance

Plume spreading was characterized using the spatial variance calculated from the normalized second-order spatial moments, centered on the centroid (see Appendix C for details). The principal longitudinal and transverse components of the plume's spatial variance, as well as its covariance (σ_{xx} , σ_{yy} and σ_{xy} , respectively), were plotted against time for all scenarios in part (c) of Figure 4-6 to Figure 4-9. These graphs offer some interesting insights into the mechanisms governing solute spreading in heterogeneous porous media.

In the longitudinal direction, σ_{xx} generally increases over time as the plume migrates and spreads out. The spreading rate is not constant, as the tracer travels through zones more or less permeable: faster areas correspond to an increased rate of spreading due to the higher

pore space velocities encountered by the migrating plume. One striking characteristic of the reversing scenario is the apparent periodical reduction in longitudinal plume dispersion, as indicated by the local negative slopes (σ_{xx} vs. t) seen in Figure 4-9(c). This indicates that the solute plume is shrinking temporarily during flow reversal periods, and lends credence to the hypothesis of a “memory effect” put forward in the preceding chapter. Note also that the transverse variance σ_{yy} initially behaves in the opposite way during the first few flow reversal events. As the experiment progresses and the plume becomes more diluted, this trend gradually becomes less apparent. The same phenomenon (plume shrinking) can be observed – albeit at a more modest scale – in the transverse direction for the zigzag scenarios. For example, the slope of σ_{yy} vs. t is negative between $t = 25$ and 30 min in Figure 4-8(c), but by a factor approximately four times smaller than the increase in longitudinal spreading σ_{xx} during the corresponding period.

Plume distortion, as portrayed by its covariance σ_{xy} , is seen to be strongly affected by both the heterogeneous nature of the porous medium, and by flow transients. Recall from Chapter 3 that σ_{xy} for the homogeneous cases was close to zero for the steady state cases, and only slightly larger for the zigzag scenarios. For the experiments reported in this chapter, however, this is only true at the very beginning of each run. In fact, σ_{xy} is sometimes observed to be of the same magnitude as the transverse variance σ_{yy} .

The mathematical model was relatively successful at reproducing the laboratory conditions, but with the important exception that longitudinal plume spreading σ_{xx} is much larger in the laboratory plume.

A first explanation may be the presence of a thin, low permeability layer against the front wall of the flow cell, designed to reduce wall effects: because the solute was initially injected across the entire thickness of the porous medium – including this low- K layer – it is likely that the resulting plume separated somewhat and migrated at two different rates. The bulk of the plume advanced faster than the slow trailing part “stuck” in the thin layer. However, numerical tests performed with parameters modified by adding such a thin layer of low permeability material (corresponding to class 9 beads) were found to produce solute distributions that were almost identical to those reported here.

A second explanation may be that chemical interactions between the tracer and the glass beads caused retardation of the plume, a process unaccounted for by the numerical models. However, laboratory batch tests using class 3 beads demonstrated that the tracer showed no affinity for sorption (see Appendix A for details). Preliminary laboratory experiments indicated that sorption could occur if the glass beads are contaminated by microbial growth after a period of stagnation. Although flow was never interrupted over the one-month period between loading the flow cell and completing all the experiments discussed in this chapter, the possibility that some microbial contamination occurred cannot be ruled out. If this was the case, then it is likely that sorption would have affected the finer beads more strongly due to their higher surface area compared to coarser materials.

2.4 Elongation and orientation

Plume elongation and orientation are shape parameters derived from second-order spatial moments. They were computed as described in Appendix C, and plotted for the current datasets – for laboratory and numerical results – in part (d) of Figure 4-6 to Figure 4-9.

For all cases, the elongation of the solute plume gradually becomes larger as the tracer is transported by the flowing groundwater. However, careful examination of the graphs shows that plume elongation can on occasion become smaller, depending on the particulars of the permeability structures it encounters and on the nature of the hydraulic gradients. Note, for example, how elongation alternately increases and decreases under the influence of reversing gradients (Figure 4-9(d)). This is consistent with the previous observation that the plume appears to shrink during flow reversal periods.

Plume orientation remains close to zero for the steady state and reversing flow scenarios (cases 9 and 12, respectively). This is true of both the laboratory and numerical datasets. On the other hand, when the hydraulic gradient direction is shifted (e.g. in the zigzag scenarios, cases 10 and 11), the orientation of the plume rotates in conjunction with the gradient. This is more apparent from the laboratory data compared to the numerical results, possibly as a result of the laboratory plume being more elongated and therefore responding to gradient direction shifts more readily.

2.5 Coefficient of skewness

Plume symmetry can be characterized by the coefficient of skewness SK , calculated from the third-order spatial moments as outlined in Appendix C. Recall that $SK \approx 0$ for a symmetrical distribution, while a negative value indicates that the data are skewed to the left (tail to the left) of the centroid; and a positive value indicates a skew to the right. Part (e) from Figure 4-6 to Figure 4-9 illustrate the temporal evolution of this parameter in both the longitudinal and transverse directions (SK_x and SK_y , respectively).

The evolution of SK_x indicates that the plume generally becomes increasingly skewed to the left. This is particularly true of the plumes observed during the laboratory experiments, but is also true of the numerical results. As discussed previously, the laboratory plumes developed a long tail which migrates slower than the bulk of the solute mass, enhancing the asymmetry and explaining the trend in SK_x vs. t .

In the transverse direction, laboratory results suggest that the plumes are significantly more symmetric than in the longitudinal direction, with SK_y generally remaining close to zero. In the numerical simulations, however, plumes exhibit a more complex symmetry history, SK_y varying between negative and positive values. Note that the transverse coordinates increase from top to bottom, which implies that a negative coefficient of skewness is expected when a plume's tail extends towards the top, and vice versa.

2.6 Kurtosis

The final parameter derived from the spatial moments is the kurtosis KR , which provides an indication of the degree of peakedness of the plumes. Calculated from the fourth-order moments (see Appendix C for details), the plume kurtosis was plotted in part (f) of Figure 4-6 to Figure 4-9. Recall that a normal distribution, $KR \approx 0$, while a negative or positive kurtosis indicates that the plume is either flatter or more peaked than normal, respectively.

In all cases, and for both the laboratory and numerical results, the initial solute distribution was essentially normal, but evolved into a slightly peaked plume. After some time, this tendency gradually disappeared as the plume began to adopt a flatter

configuration. The kurtosis occasionally reached values of -1.2 or less, indicative of bi- or multi-modal behaviour.

3 MACRODISPERSIVITY

In order to characterize the overall evolution of the plume spreading, macrodispersivity A_{ij} was calculated with the *Global* and *Absolute Stepwise* methods, as in the previous chapter. The details about the calculation methods are presented in Appendix D.

3.1 Confidence intervals

The vertical error bars in the graphs presented in the following section provide an estimate of the uncertainty associated with the experimental results. The procedure used to calculate the confidence intervals (*CI*) is almost identical to that used in the previous chapter, and is described in detail in Appendix D.

The macrodispersivity was obtained from the slopes of $\bar{v}_i$ vs. t and D_{ij} vs. t , where i refers to the x and y directions. The regression standard errors on the slope (*SE*) were used to characterize the uncertainty resulting from each slope estimate. These error estimates were then combined using the superposition-of-error theorem [Leaver and Thomas, 1974] when calculating the derived quantities ($\|\mathbf{v}\|$, A_i and A_x/A_y). However, the precision error could not be determined as was done in Chapter 3, because no replicate runs were available for the experiments reported in this chapter. Instead, the precision error was assumed to be the same as for the homogeneous case.

3.2 Macrodispersivity results

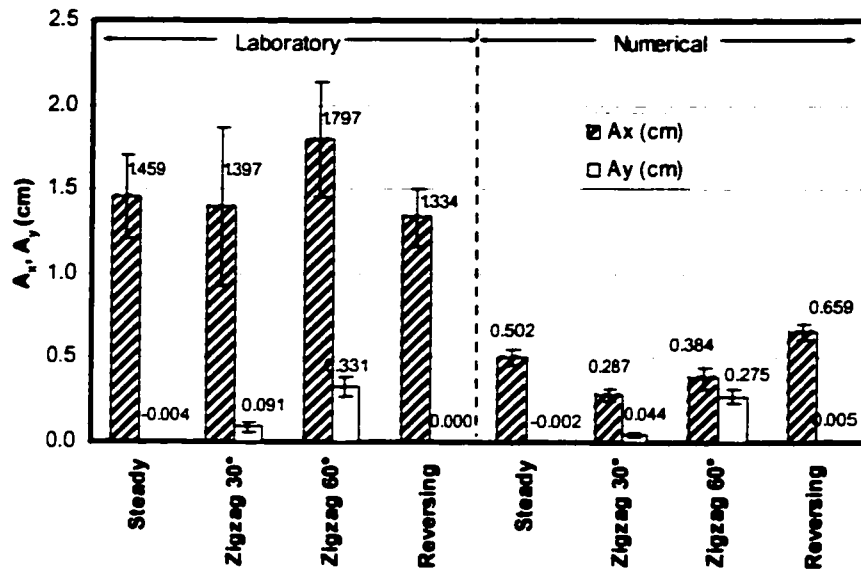
The macrodispersivity estimates derived from the heterogeneous experiments presented in this chapter are not as easily interpreted as those reported of the homogeneous cases. In particular, the trends between scenarios are not quite as clear as they were for the simpler system of Chapter 3. This is probably explained in most part by the non-ideal nature of the heterogeneous porous medium. The principal results from the laboratory and numerical experiments are summarized below using a graphical representation. These include

longitudinal and transverse macrodispersivity (A_x and A_y , respectively), estimated using both the *Global* (Figure 4-10(a)) and *Absolute Stepwise* (Figure 4-10(b)) methods (refer to Appendix D for a discussion of the methods). Figure 4-11 presents the ratio of the transverse to longitudinal macrodispersivity (A_x/A_y), while estimates of the magnitude of the velocity vector ($\|v\|$) calculated using these two methods are presented in Figure 4-12. A number of observations about these data are outlined here.

Figure 4-10(a), shows the macrodispersivity estimated for all scenarios, including both laboratory and numerical results, and calculated using the *Global* method. In all cases, A_x is significantly larger than A_y , for laboratory and numerical data. An obvious difference in the laboratory compared to the numerical results is that A_x and, to a smaller extent, A_y , are both smaller in the former. Additionally, laboratory results exhibit a higher uncertainty. Flow direction variations increase A_y in proportion to the angle of rotation, while no trend is apparent for A_x . Finally, we see that A_y is almost zero for both the steady state and reversing cases, and the difference in A_x is marginal when comparing the two scenarios.

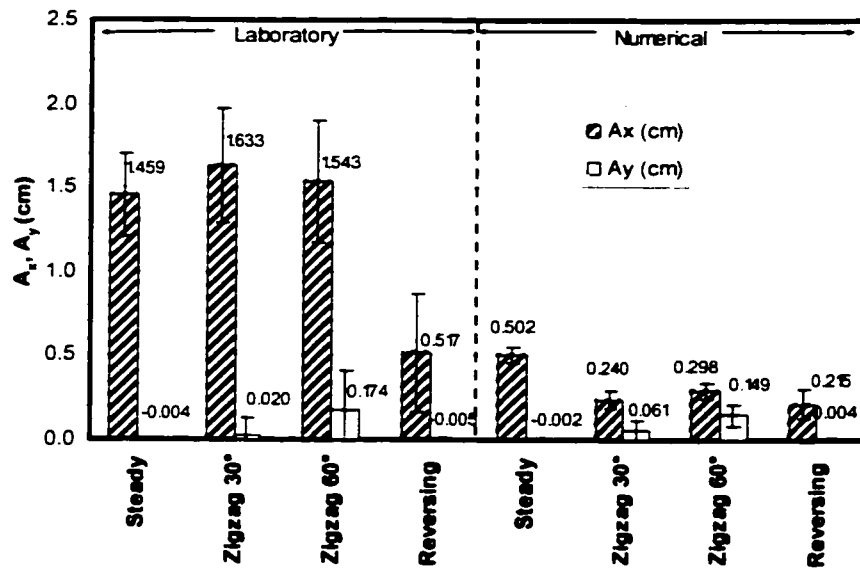
Results calculated from the *Absolute Stepwise* method, shown in Figure 4-10(b), show a few differences with their *Global* counterparts, discussed in the previous paragraph. First, for the reversing case, A_x derived from both the laboratory and numerical experiments is significantly smaller compared to the *Global* results. Also, the uncertainty appears to be slightly larger in estimates obtained using the *Absolute Stepwise* method. Moreover, variations in the direction of flow increase A_y proportionally to the rotation angle, although the increase is smaller than what was seen in the *Global* results.

**Longitudinal and Transverse Macrodispersivity
(Laboratory & Numer. Results - Global Method)**



(a)

**Longitudinal and Transverse Macrodispersivity
(Laboratory & Numer. Results - Abs. Stepwise Method)**



(b)

Figure 4-10. Longitudinal and transverse macrodispersivity (A_x and A_y , respectively) estimated from laboratory experiments and numerical simulations using the *Global* and *Absolute Stepwise* methods. Error bars indicate the 95% confidence interval.

The relative sizes between the transverse and longitudinal components of macrodispersivity are compared in Figure 4-11, showing for all scenarios the results calculated from the laboratory and numerical experiments. First, the ratio A_y/A_x is close to zero for both the steady and reversing cases because of the small magnitude of A_y ; this is seen in both the laboratory and numerical results. Comparing A_y/A_x between data derived from the laboratory and numerical results reveals that the relative importance of A_y is much greater in the latter, and that this discrepancy becomes more important as the rotation angle increases. Finally, despite the large uncertainty, there appears to be a slight difference between the ratio A_y/A_x calculated using the *Global* method, as opposed to the *Absolute Stepwise* method; the former produces results that are slightly larger in most cases, the only exception being the “Zigzag 30°” case (numerical results only), where the opposite relationship is observed.

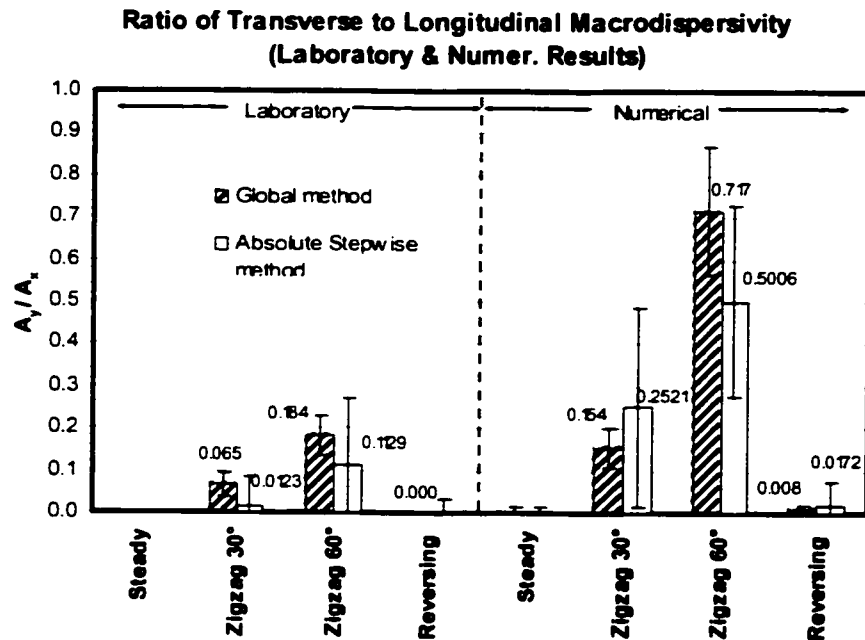


Figure 4-11. Ratio of the transverse to longitudinal macrodispersivity (A_y/A_x), calculated using the *Global* and *Absolute Stepwise* methods from laboratory and numerical results. Error bars indicate the 95% confidence interval.

The magnitude of the velocity vector, estimated for all heterogeneous experiments using the *Global* and *Absolute Stepwise* method, is plotted in Figure 4-12. Two characteristics of this graph are worth noticing. First, the laboratory estimates of $\|\mathbf{v}\|$ are significantly smaller than their numerical counterparts. This is true for all cases, regardless of the calculation method used. Second, significant differences can be seen between the *Global* and *Absolute Stepwise* methods when calculating velocity, based on the nature of the flow direction variations. In the former method, $\|\mathbf{v}\|$ decreases as the gradient rotation angle increases, while for the *Absolute Stepwise* method velocities are relatively uniform across scenarios.

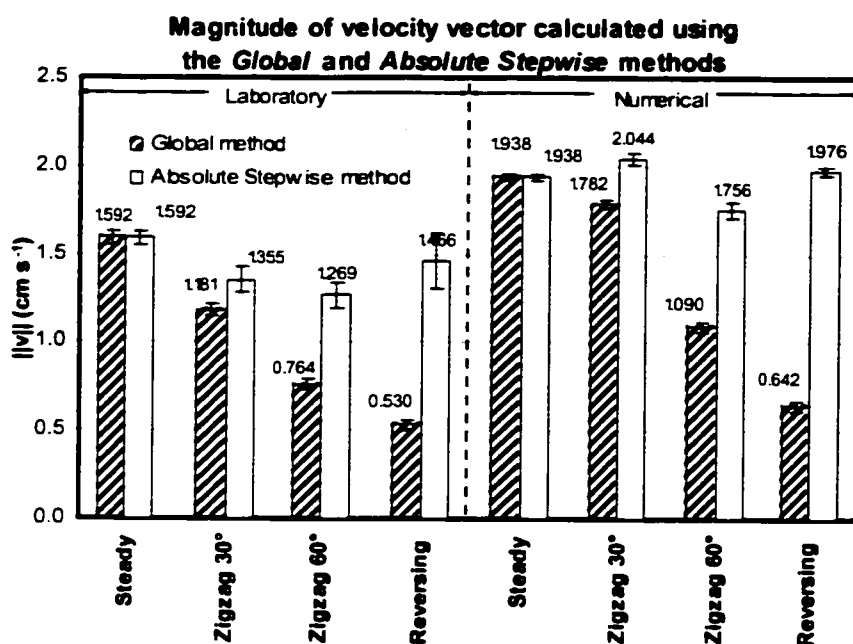


Figure 4-12. Magnitude of the velocity vector $\|\mathbf{v}\|$ calculated using the *Global* and *Absolute Stepwise* methods. Results are shown for both the laboratory and numerical datasets. 95% confidence intervals are indicated by the vertical error bars.

The results presented in Figure 4-10 to Figure 4-12 are discussed in more detail below, with particular emphasis on (1) the effect of the hydraulic gradient direction (flow pattern), (2) the effect of the calculation method, and (3) a comparison of laboratory vs. numerical results.

3.3 Effect of the hydraulic gradient direction (flow pattern)

Four scenarios were investigated using the heterogeneous porous medium, to assess the influence on plume migration of variations in the direction of the hydraulic gradient. This allowed for a comparison between macrodispersivity estimates resulting from the transient scenarios (cases 10 to 12) to results from the steady state scenario (case 9).

The magnitude of the velocity vector $\|\mathbf{v}\|$ estimated using the *Absolute Stepwise* method exhibits small but significant variations across the flow scenarios (see Figure 4-12). Because the magnitude of the hydraulic gradient was identical for all cases, these velocity variations result entirely from the heterogeneous nature of the hydraulic conductivity field. Note that the velocity from the reversing scenario is almost identical to that of the steady case. This is not surprising because in both cases the plume travels through an identical portion of the porous medium, and therefore “experiences” the same level of heterogeneity. For the zigzag scenarios (cases 10 and 11), the fact that $\|\mathbf{v}\|$ is smaller suggests that the plume migrated across areas of lower permeability. If, on the other hand, the *Global* method is used to calculate $\|\mathbf{v}\|$, the apparent velocity can be severely underestimated if the changes in flow direction are important.

Variations in gradient direction significantly affect plume spreading, in particular in the transverse direction. Figure 4-10 shows the longitudinal (A_x) and transverse (A_y) macrodispersivity estimates, calculated from laboratory and numerical results using the *Global* and *Absolute Stepwise* methods. Comparing A_x (shaded data series) across scenarios shows some variations, but does not suggest any trends. Contrary to what we saw in the previous chapter for a simpler (homogeneous) system, gradient variations in this case do not appear to influence longitudinal plume spreading in a systematic manner. In the transverse direction, however, shifting the gradient direction over time enhances the spreading rate. Whereas A_y is negligible in the steady and reversing scenarios, it increases significantly as the transverse gradient increases during flow shifts. Moreover, this effect – which can be seen from the laboratory and numerical results and independently of the calculation method – is proportional to the rotation angle. While both zigzag flow scenarios enhance A_y , this increase is much more important for gradient shifts of 60° compared to 30°. This can also be

seen by looking at the ratio A_y/A_x , shown for the laboratory and numerical results in Figure 4-11. For the steady and reversing cases this ratio is essentially zero. On the other hand, the effect on A_y of introducing direction shifts in the hydraulic gradient (zigzag cases) increases the ratio very quickly.

3.4 Effect of the calculation method

As was discussed in the previous chapter, selection of the calculation method has a dramatic impact on the estimates of $\|\mathbf{v}\|$ or A_i . Looking at the above graphs (Figure 4-10 to Figure 4-12), one can see a number of striking differences between *Global* and *Absolute Stepwise* results. These differences are explained in good part by the fact that the *Global* method, as its name implies, “looks” at the plume evolution globally, without consideration for the nature of the hydraulic gradient. The second method, on the other hand, explicitly combines estimates from a number of time intervals over which the gradient imposed on the system remained constant.

The most obvious difference between the calculation methods is the underestimation by the *Global* method of the magnitude of the plume velocity. In fact, for any given scenario, the discrepancy between the *Global* and *Absolute Stepwise* estimates ($\|\mathbf{v}\|_G$ and $\|\mathbf{v}\|_{AS}$, respectively) corresponds approximately to what can be estimated by looking at the nature of the gradient direction changes. For instance, $\|\mathbf{v}\|_G / \|\mathbf{v}\|_{AS} = 0.530 / 1.466 = 0.36$ for the reversing scenario (case 12). This is because the forward flow periods last twice as long as the reversing events, resulting in a “net” globally averaged gradient approximately equal to one third of its absolute magnitude. For the zigzag scenarios (cases 10 and 11), the *Absolute Stepwise* and *Global* estimates differ by a factor roughly equivalent to $\cos(30^\circ)$ or $\cos(60^\circ)$, respectively. These observations are identical to those discussed in the previous chapter for the homogeneous porous medium.

Macrodispersivity estimated from the steady state results are identical regardless of the calculation method used. A number of differences can be seen for the transient cases, however. Moreover, A_x and A_y both seem to be overestimated by the *Global* method compared to estimates obtained from the *Absolute Stepwise* method. This is especially true

of the A_x estimate from the reversing case. This is explained by the fact that A_{ij} is inversely proportional to $\|\mathbf{v}\|$.

3.5 Laboratory vs. numerical results

The following observations can be made comparing results from numerical simulations to those from laboratory experiments:

1. Figure 4-12 clearly shows that $\|\mathbf{v}\|$ derived from the numerical results is consistently larger than in the laboratory results, for any given scenario and regardless of the calculation method;
2. The laboratory experiments produced A_x and A_y estimates that were much larger than in numerical simulations (see Figure 4-10(a) and (b)); and
3. The relative magnitude of transverse compared to longitudinal macrodispersivity, portrayed by the ratio A_y/A_x in Figure 4-11, was larger in numerical simulations than in the laboratory model

CHAPTER 5 – Summary and Conclusions

Science is built up of facts, as a house is built of stones; but an accumulation of facts is no more a science than a heap of stones is a house.

– Henri Poincaré

No amount of experimentation can ever prove me right; a single experiment can prove me wrong.

– Albert Einstein

The main focus of this research was to study the effect of transient flow on contaminant dispersion in porous media. This was addressed using physical and numerical models, and involved both homogeneous and heterogeneous porous media. The dispersive behaviour of dissolved contaminant plumes travelling through these systems was monitored through the spatial moments. The effects of deterministic flow transients were assessed quantitatively by comparing the macrodispersivity calculated from flow scenarios involving steady state and transient gradients. By controlling the magnitude and direction of the hydraulic gradient, 12 distinct scenarios were investigated; eight in a homogeneous porous medium and four in a heterogeneous one. The results from the homogeneous experiments and numerical simulations were presented in Chapter 3; and those of the heterogeneous scenarios in Chapter 4.

This chapter summarizes these findings and outlines the main conclusions that can be drawn from this study; a few comments are provided discussing the limitations of some aspects of the experimental approach; some possible directions that could be explored in the footsteps of this study are also discussed; finally, some of the practical implications of this work are discussed.

Recall from Chapter 1 the principal questions that were identified at the onset of the study, namely:

1. Is it possible to reproduce in the laboratory the complex hydraulic conditions encountered in nature, in particular by combining flow transients with heterogeneous porous media ?
2. Is it possible to accurately monitor, using a physical model, the spatial distribution and temporal evolution of a contaminant plume migrating through homogeneous or heterogeneous porous media ?
3. What are the impacts on macroscopic dispersion of transience in the hydraulic gradient, both in terms of its magnitude and its direction, and how does this compare with previously published results ?
4. Do flow transients influence contaminant dispersion differently in heterogeneous compared to homogeneous porous media ?
5. Can the commonly used numerical models MODFLOW and MT3D reproduce the experimental results obtained during laboratory experiments under controlled steady and transient flow conditions ?

The methods used to address these questions were outlined in Chapter 2 and discussed in detail in several appendices. The principal observations made from the analysis of the experimental evidence are summarized and discussed below.

1 EXPERIMENTAL APPROACH AND TECHNICAL INNOVATIONS

The innovative design of the laboratory model and peripheral apparatus proved to be satisfactory in addressing all of the above questions. In particular, the experimental design was shown to be capable of (1) reproducing in the laboratory a heterogeneous porous medium with a predetermined permeability structure, and (2) imposing deterministic transients on the hydraulic gradient. Additionally, the spatial moment analysis using digital photographs to monitor plume evolution showed that it is possible to accurately track the spatial and temporal evolution of a dissolved contaminant plume. The most significant

contributions of this work, in terms of the experimental methods that were used, are summarized below.

First, the hydraulic control system (HCS) allows for instantaneous changes in flow direction – or even reversals – without the need to interrupt the experiments or otherwise disturb the flow field. An additional useful feature of the HCS is to allow flow to be instantly stopped as required. For example, this was done at the beginning of each experiment during tracer injection, to minimize smearing; and to enable a reproducible release into the flow cell of a compact initial plume.

Second, the front-loading design of the flow cell was successful in allowing the emplacement of heterogeneous porous media of almost any prescribed permeability structure. The complexity of the porous medium that can be created using this model is limited on the one hand by the size and shape of the emplacement template's compartments... and on the other by the experimenter's patience during the labour-intensive process of loading the granular materials into each compartment ! This unique advantage of the flow cell was illustrated with the heterogeneous porous medium introduced in Chapter 2 and described in detail in Appendix A. The results of the experiments conducted in this spatially variable, randomly correlated hydraulic conductivity field are discussed in Chapter 4. Additionally, we found useful the ability to adjust the thickness of the porous medium between 3 and 5 cm. This tolerance allowed flexibility that was necessary to optimize the system, given the physical constraints of the length of the inlet/outlet ports, the height of the lateral wall and the thickness of the cover.

Third, due to the ability of the system to control the flow direction, particularly in the reversing flow experiments, it was possible to examine indirectly the wall effects caused by the increased porosity at the interface between the porous medium and smooth walls of the flow cell. The homogeneous experiments (Chapter 3) under reversing flow conditions demonstrated how wall effects can allow a thin portion of the contaminant plume to advance at a faster rate compared to the bulk movement of the plume. These observations were made from the temporal evolution of the parameters derived from the spatial moments, in particular the total mass, variance and coefficient of skewness.

Fourth, we clearly showed that by selecting the tracer and camera filter carefully, and by processing the acquired images using an appropriate sequence of operations, it is possible to isolate and fully characterize a tracer plume, even in an optically non-uniform porous medium. The spatially variable brightness of the heterogeneous system introduced additional technical challenges that made the interpretation of these results somewhat more difficult than in the case of a homogeneous system. Nevertheless, the techniques presented in this work provide a valuable tool for observing in real time solute migration in heterogeneous porous media. The evolution of the plume, whose distribution is integrated over its thickness in the direction normal to the images, can then be followed as it migrates through the system.

Fifth, the quantitative procedure of using spatial moments derived from images as experimental evidence for tracking the migration of contaminant plumes, is the first of its kind. The high-resolution images had the advantage of producing a continuous concentration distribution in space and discrete in time, without disturbing or interrupting flow. Neither using spatial moments to characterize a contaminant plume, nor tracking the evolution of a tracer from transmitted light images are original ideas as such. To our knowledge, however, the fact that these two techniques were used in conjunction constitutes an innovative approach. Moreover, the plugin developed during this study, to compute the spatial moments from a contaminant plume monitored via an image sequence, was made available to the scientific community (see Appendix C for details).

2 FUTURE IMPROVEMENTS TO THE EXPERIMENTAL DESIGN

The main challenge that was faced in order to lead this investigation to a successful completion was to design and develop the apparatus necessary to impose flow transients in the laboratory. A close second was designing a flow cell that allowed for the easy construction of heterogeneous porous media with prescribed permeability structures. Despite many frustrating obstacles and failed preliminary attempts, it was with great satisfaction – and admittedly, pride – that the hydraulic control system and front-loading flow cell introduced in this work were finally completed. Of course, the combination of techniques

used to acquire, process and interpret the experimental results is not without limitations, some of which are discussed in the following section.

A number of issues were raised throughout this study in relation to a number of limitations related to the experimental design. In particular, some concerns were raised in accompanying chapters and appendices about (1) the image calibration procedure, (2) the influence of the wall effect, (3) the restrictions imposed on the correlation lengths, and (4) the ability to modify the magnitude of the hydraulic gradient rapidly. These limitations are summarized below and are accompanied by some recommendations that might be applied to help circumvent or improve upon them.

2.1 Image calibration

Calibrating images to convert pixel grey levels into actual solute concentrations posed a significant challenge. Our initial approach was to saturate the central portion of the flow cell with a solution of known concentration to produce one calibration image for each of several calibration solutions of a different concentration. This produced a linear relationship between the normalized pixel values (G) averaged over the dye-saturated area, and the known concentration of the solutions (C_{cal}). Unfortunately, the G vs. C_{cal} calibration curve proved to be useless for calibrating images containing solute plumes. The concentration near the centre of the plume immediately following injection was found to significantly over-predict the “true” value (which should have been close to the known concentration that was injected). The exact cause for this discrepancy is not clearly understood at this point. We suspect, however, that the calibration error might result from an alteration of the grey levels measured at each pixel location within the plume, caused by a “bleeding” of the relative bright surrounding pixels, where the tracer is absent.

2.2 Wall effect

The increased permeability at the interface between the porous medium and the flat surfaces of the cell’s bottom and front panels was shown to significantly alter the migration of the dissolved tracer. This artefact of the laboratory design is a phenomenon referred to as the

“wall effect”. Appendix F discusses this process in greater detail, using the reversing flow experiments conducted in a homogeneous medium to illustrate how the wall effect causes the creation of a “fast” plume. This can negatively impact laboratory tracer experiments that typically observe the tracer migration in the top few millimetres immediately adjacent to the walls [e.g. *Schincariol and Schwartz*, 1990]. In such cases, the plume velocity might be over-predicted because the solute distribution in fact monitors the fast plume rather than the actual (slower) plume that travels at the same velocity as the groundwater. The impact of the wall effect in the present study is thought to be relatively unimportant, however, because the concentration distributions were obtained from transmitted light images, providing an integrated view of the contaminant plumes.

The wall effect can be minimized by ensuring that the porous medium is tightly compacted inside the smooth-walled flow cell. Gluing the granular materials onto the wall surfaces and increasing the thickness of the porous medium can also reduce this phenomenon [*Bear*, 1972]. Altering the flow cell’s inside surfaces to render them rougher, possibly by sand blasting, might also help to further reduce the wall effect. Care must be exerted, however, in ensuring that the strategy adopted to circumvent this problem does not alter the optical properties of the flow cell. This consideration is important if, for example, images are to be used to monitor the tracer migration, but may not be relevant if sampling ports or electrical probes are used to measure the solute concentration distribution.

2.3 Correlation scales

The ability to recreate spatially variable porous media in the laboratory can be limited by the physical restrictions of the laboratory setup. In Chapter 4, we discussed the constraints imposed on the selection of the correlation scales. First, the correlation scale must be small enough given the dimensions of the flow domain to ensure that the requirements for ergodicity are met (the flow domain should include at least 10 correlation scales in every direction). At the same time, we must select correlation scales that are at least four times larger than the grid spacing used to create the porous medium, to reproduce the desired degree of heterogeneity with a high enough resolution. The physical restrictions of the laboratory apparatus limited our flexibility in choosing correlation lengths. With a flow cell

size of only 65 cm and the relatively large template compartments (1.5 cm), we were forced to use an isotropic correlation length equal to 6.0 cm. To use other correlation lengths, for example in the construction of anisotropic heterogeneous porous media, would be possible only if a template with smaller compartments and/or a larger flow cell is/are used.

2.4 Gradient magnitude variations

Although the hydraulic control system allowed for variations in the flow magnitude, the time required when adjusting the hydraulic gradient limited our ability to apply this type of flow transience rapidly. As described in Appendix A, the magnitude of the gradient was modified by altering the tilt angle of the overflow manifold, which caused the hydraulic head in each input/output reservoir to change accordingly. Although this modification takes place almost instantaneously, it must be accompanied by a manual adjustment of the supply of fresh water into the upgradient (input) reservoirs. This rendered impractical real-time variations in the flow magnitude. Designing a new replenishment system that would permit a quick (or ideally, automatic) adjustment of the supply rate in each channel would provide the functionality necessary to study flow scenarios involving variations in gradient magnitude.

3 IMPACT OF FLOW TRANSIENCE ON CONTAMINANT DISPERSION

Changes in the mean flow direction have a significant impact on the spreading rate of contaminant plumes migrating through homogeneous and heterogeneous porous media. In particular, transverse macrodispersivity (A_T) was shown to increase in proportion to the rotation angle in mean hydraulic gradient, while the longitudinal component (A_L) decreased, but by a smaller amount. This is in line with the findings of *Kinzelbach and Ackerer [1986]* and *Goode and Konikow [1990]*, which suggested that an increase in apparent transverse dispersion due to flow transients had to be accompanied by a corresponding reduction in apparent longitudinal dispersivity in advection-dominated problems. Note that contrary to the homogeneous case where rotations in the gradient clearly cause A_L to decrease, no systematic trend was apparent for the heterogeneous porous media. This indicates that the

spatial variability can mask the temporal variations in the flow field, even when flow transients are important.

During the laboratory experiments, where solute plumes were subjected to periodically reversing flow conditions, the longitudinal spreading rate was occasionally observed to decrease during flow reversals, suggesting that the plume was actually shrinking temporarily. This surprising behaviour, observed in the homogeneous and heterogeneous experiments, seems to contradict Bear [1972] regarding the irreversibility of the dispersion process. If the dispersion process were truly irreversible, one would expect plume spreading to only increase over time, even during flow reversal events. Needless to say, however, that reversing the flow will not return the tracer distribution to its initial configuration, although results of this work have shown that this is true of at least a portion of the dissolved plume. The concept of “memory effect” was introduced in Chapter 3 as a possible interpretation for this surprising behaviour. To illustrate this, consider the case where a plume is subjected to periodically reversing gradients, but with net forward flow (“forward” intervals last longer than “reverse” intervals). The memory effect causes the dissolved solutes to encounter the same realization of a portion of the aquifer three times: once initially during forward flow, a second time after flow is reversed, and finally when the forward flow conditions are resumed. Presumably, this induces a correlation each time the solute travels through the same region, thereby reducing the randomness of the trajectory it can follow. No medium is truly homogeneous in space at the pore scale, and it is likely that dissolved solutes travel for the most part through the pathways that are most permeable, following tortuous trajectories through geologic materials along paths of least resistance. The complex arrangement of branching conduits may result in hydraulic head distributions that are somewhat different when the bulk hydraulic gradient is reversed compared to its pre-reversal configuration.

Selection of the calculation method to characterize the contaminant plume’s dispersive behaviour can have a considerable influence on the results, particularly when the flow transients are important. This is true regardless of the nature of the porous medium. Appendix D presents four calculation methods that can be used to derive macrodispersivity A_{ij} . These methods were identified based on the selection of the time interval(s) to estimate $\bar{v}_i$ and D_{ij} from the slopes x_c vs. t and σ_{ij} vs. t , respectively. The *Absolute Stepwise* method,

by forcing all slope segments to be positive, was shown to produce macrodispersivity estimates that were significantly different from those obtained using other methods.

An argument can be made that the *Absolute Stepwise* method is the most appropriate of the methods used here. Another approach that may be considered in future work would be to take the positive root mean square of the velocities (quadratic mean) instead of the absolute value. The application of any of the stepwise methods requires water-level data to be collected frequently to capture the flow transients. Consider a scenario where the hydraulic gradient is reversed periodically, but where the forward and reverse intervals are of equal duration (this might be representative, for example, of an aquifer influenced by tides). After an even number of forward-reverse cycles, the plume's centre of mass would return to its initial location, with a net travelled distance close to zero. The resulting apparent velocity would also be near zero, while the plume would have dispersed noticeably, having experienced real movement through the porous medium during the experiment. Clearly, the *Absolute Stepwise* method will provide a more realistic evaluation of the plume's true spreading rate – or macrodispersivity. The small velocity obtained from the other three methods would overestimate A_{ij} by ignoring the contribution of flow transients to dispersion. Conversely, the “memory effect” would be most apparent in this scenario, which would result in an overestimation of the macrodispersivity by the *Absolute Stepwise* method. The *Absolute Stepwise* method requires that the flow conditions be quantified with a sufficient temporal resolution in order to fully incorporate the flow transients into the calculations. This is not always possible in field studies, where water levels are typically monitored using a frequency and/or spatial density that may not allow flow transients to be recognized or fully characterized.

As expected, the complex nature of the heterogeneous porous medium causes migrating contaminant plumes to be more highly dispersed and distorted than when the porous medium is uniform. This is not surprising when considering the increased variability of pore-scale velocities caused by the spatial variations in hydraulic conductivity. This is true for both A_y and, to a lesser extent, A_x regardless of the calculation method used. Also, the ratio A_y/A_x is much smaller for plumes migrating through heterogeneous than in

homogeneous media, and appears to be due principally to the spatial variability in the porous medium.

The degree of agreement between macrodispersivity obtained from numerical simulations compared to estimates derived from laboratory experiments depended on whether the porous medium was homogeneous or heterogeneous. For the more idealized homogeneous porous medium (Chapter 3), the agreement was relatively good. This is not surprising given the tightly controlled laboratory conditions used in these experiments. Conversely, the results in a heterogeneous porous medium resulted in several noticeable differences. Specifically, the laboratory model produced lower velocity and higher macrodispersivity estimates than predicted using the numerical model. The source of the discrepancy between physical and numerical models most likely results from an inability of the numerical model to completely reproduce the experimental conditions.

4 FUTURE DIRECTIONS

The findings outlined and discussed in this study give rise to several possibilities for the advancement of the current state of knowledge of the behaviour of contaminants in the subsurface. The most significant features of the laboratory apparatus, namely to allow flow transients to be easily imposed on the system, and to permit the construction of a wide range of permeability structures, open the door to address some important questions. Examples of interesting topics that could be investigated in the future include:

1. Performing statistical analyses of the effects of flow transients on contaminant dispersion using multiple realizations of stochastically-generated permeability fields. This laboratory Monte Carlo approach is a logical extension of the work presented in the current investigation. This type of study is commonly done using numerical models, by imposing an identical set of flow conditions using several realizations of a hydraulic conductivity field [e.g. *Naff et al.*, 1998b; *Naff et al.*, 1998a]. The experimental apparatus introduced in this study provides an opportunity to conduct similar studies to elucidate inadequacies in the numerical models in multiple-realization studies.

2. Assess the influence on plume migration of various configurations or structures in the hydraulic conductivity distribution. The heterogeneous porous medium studied in this work consisted of a statistically isotropic random field of prescribed mean, spatial variance and correlation lengths. The front-loading flow cell presented here is not limited to this type of system, and the packing procedure described in Appendix A could easily be adapted to produce virtually any type of permeability structure. This might include, for example, the creation of permeability contrasts in the form of lenses of various sizes and shapes, a layered system, or any other similar structure requiring an organized, controlled arrangement of granular materials. Additionally, one could generate an anisotropic or layered system where the hydraulic gradient might be applied parallel, perpendicular or at other angles relative to the principal axes of the hydraulic conductivity field.
3. Investigate further the “memory effect”. It was apparent from laboratory experiments in a simple, somewhat idealized homogeneous medium that reversing the hydraulic gradient reduces the randomness of the mixing process; this induced correlation leads to a reduced apparent macrodispersivity compared to an equivalent steady state scenario. In particular, it was impossible, using the transmitted light experimental model and image processing techniques used in this work, to determine with certainty whether the apparent plume shrinkage was simply an artefact of the wall effect, or whether it was the true manifestation of an induced correlation, or “memory effect”. Conducting a forced-gradient field investigation, where reversing flow conditions could be imposed on a natural porous medium, would provide an opportunity to gain further insight into this memory effect.

5 PRACTICAL IMPLICATIONS

The main impact on dissolved contaminant plumes of flow transients was shown to be the enhanced dispersion, particularly in the direction transverse to the mean gradient, caused by the additional variability in the flow field. The increased dispersion is in itself beneficial to

the natural attenuation of the contaminant plume. By extension, the increased volume occupied by the plume due to the increased spreading can have a positive impact on most other natural attenuation processes as well; these include biodegradation, dilution, sorption, volatilization, radioactive decay, and chemical or biological stabilization, transformation, or destruction of contaminants [National Research Council, 1997].

The differences observed within a single experimental dataset between macrodispersivity estimates obtained using different calculation methods will hopefully serve to caution other researchers when using spatial moments to characterize plume spreading. In field, laboratory or numerical studies where flow transients exist (or are suspected to exist), one should pay particular attention to the selection of the time interval(s) used to estimate the time rate of change of $\bar{v}_i$ and D_{ij} . For example, in situations where a contaminant plume is subjected to reversing hydraulic gradients, bulk velocity estimated using the *Global* method will be underestimated due to the contribution of negative $\bar{v}_x$ values during flow reversal periods. This will in turn result in an overestimation of the true dispersive potential, or macrodispersivity, of the porous medium.

The significant differences between results derived from laboratory compared to numerical models raised some concerns regarding the ability of the numerical model to capture the full complexity of the combined contribution to hydrodynamic dispersion of spatial and temporal variability. We demonstrated earlier that the uncertainty associated with the estimation of dispersion is an important limiting factor in our ability to predict plume migration with accuracy. In the context where natural attenuation mechanisms are used as a remedial strategy, mathematical models are often an integral part of feasibility studies required in order to assess the applicability of monitored natural attenuation (MNA) at a given site. Consequently, the detailed characterization performed in support of MNA to remediate a contaminated site should necessarily include an evaluation of the nature and importance of flow transients. This might imply, for example, increasing the frequency of water level data collection, particularly during the initial stages of a field study.

Finally, the results of this work might be useful in improving existing numerical models. For instance, imposing controlled flow transients to a large number of heterogeneous

systems might help in the development of a mathematical formulation that would incorporate the effects of flow transience into estimates of macroscopic dispersion derived from field results, but without an explicit characterization of the changes of hydraulic heads over time. It is possible, for example, that the scale-dependent dispersive behaviour observed in large contaminant plumes [e.g. *Gelhar et al.*, 1992] can be partly explained by flow transience which is usually neglected, and therefore not explicitly accounted for in the dispersion estimates. The concept of “dispersivity” as it applies to numerical simulations of solute transport is meant to account for the spatial heterogeneities that occur at scales smaller than the representative elementary volume. It should logically be a function of the porous medium alone and should not change with the problem scale. It is, however, necessary to adjust this parameter (usually by calibrating the numerical model to field measures of solute concentrations) in order to compensate for this apparent scaling effect. Presumably, being able to incorporate a measure of the flow transience into the models might help solve this discrepancy.

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APPENDIX A – Laboratory Apparatus

“The sand box model (granular or physical model) is a reduced scale representation of the natural porous medium domain. It is a true model in the sense that both prototype and model involve flow through porous media”
[Bear, 1972, p. 678]

– Jacob Bear

This appendix describes the components that make up the experimental setup. First, the physical model’s flow system is presented, including the flow cell and peripheral apparatus used for flow control. Second, the physical properties of the porous media are described in detail along with their emplacement methods. Third, the tracer properties are discussed. Finally, the image acquisition system used to characterize the tracer concentration distributions is described and discussed.

1 PHYSICAL MODEL

A significant contribution of this study was the design of a physical model for simulating changing flow directions in porous media. A key requirement was that the flow control system had to allow the mean flow direction and/or magnitude to be modified rapidly during the course of a solute transport experiment, with minimal transitional flow field instabilities between changing flow conditions. This section provides the technical details of the setup of both the flow cell itself, as well as the peripheral apparatus used for controlling flow in the model.

Figure A-1 provides an overview of the various components of the experimental setup, described in the following sections.

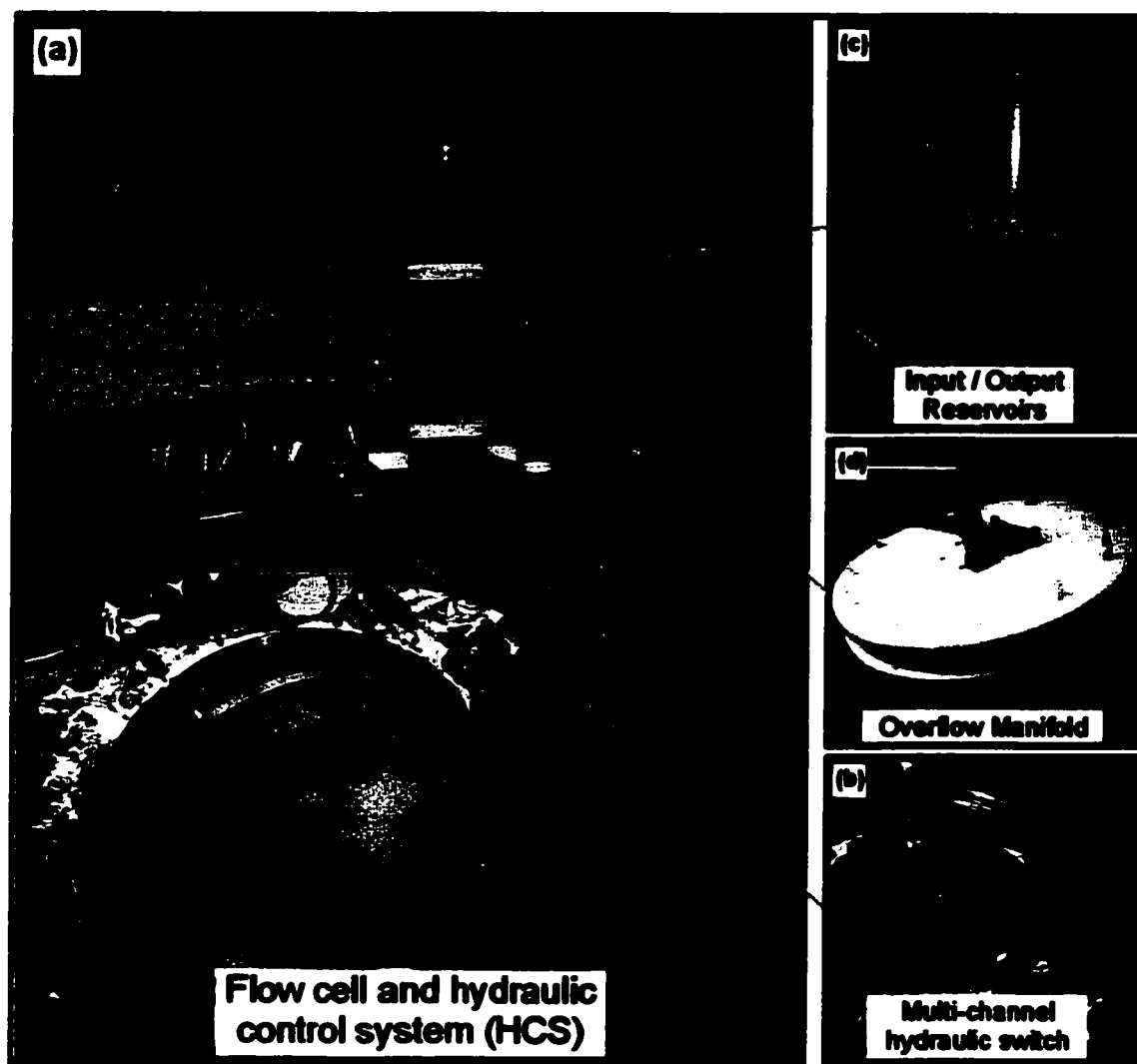


Figure A-1. Experimental apparatus, including a global view of the physical model (a), as well as close-ups of some of the key components of the hydraulic control system (HCS): (b) the multi-channel hydraulic switch, (c) the inlet / outlet reservoirs, and (d) the overflow manifold. Distilled water was supplied to the upgradient (input) reservoirs using a fixed head tank placed directly above the reservoir support rack.

1.1 Flow cell

The laboratory experiments were performed using a circular disk-shaped clear acrylic flow cell, shown schematically in Figure A-2. The internal diameter of the flow cell was 65 cm, and its adjustable thickness between 3 and 5 cm. Although the flow cell was circular, the arrangement of injection and extraction ports around the cell produced groundwater flow

fields that were essentially unidirectional at any given time, with equipotential lines being parallel to each other. The unconventional circular shape was selected to minimize flow cell geometry effects in experiments where the mean groundwater flow direction was rotated. Relatively thick (2.54 cm) acrylic (Plexiglas[®]) was used in the construction of the top and bottom panels. This was necessary in order to eliminate possible distortions due to a variable thickness of the porous medium, while preserving the high translucence of the flow cell. The 0.635 cm thick lateral wall and supporting flange (4.76 cm wide by 1.27 cm thick) were made of polypropylene (PP). The somewhat flexible PP flange was secured onto the bottom Plexiglas[®] panel, and was pressed down using a 0.635 cm thick aluminum “ring”. A 0.24 cm thick Neoprene gasket was sandwiched between the PP flange and the bottom acrylic panel to provide a watertight seal. The top panel (cover) was designed to be removable to allow topside loading and packing of the porous medium into the cell. The top acrylic panel was machined to fit precisely inside the lateral wall and a pair of o-rings placed along the circumference of the cover, allowing the top panel to be easily removed, while effectively sealing the flow chamber.

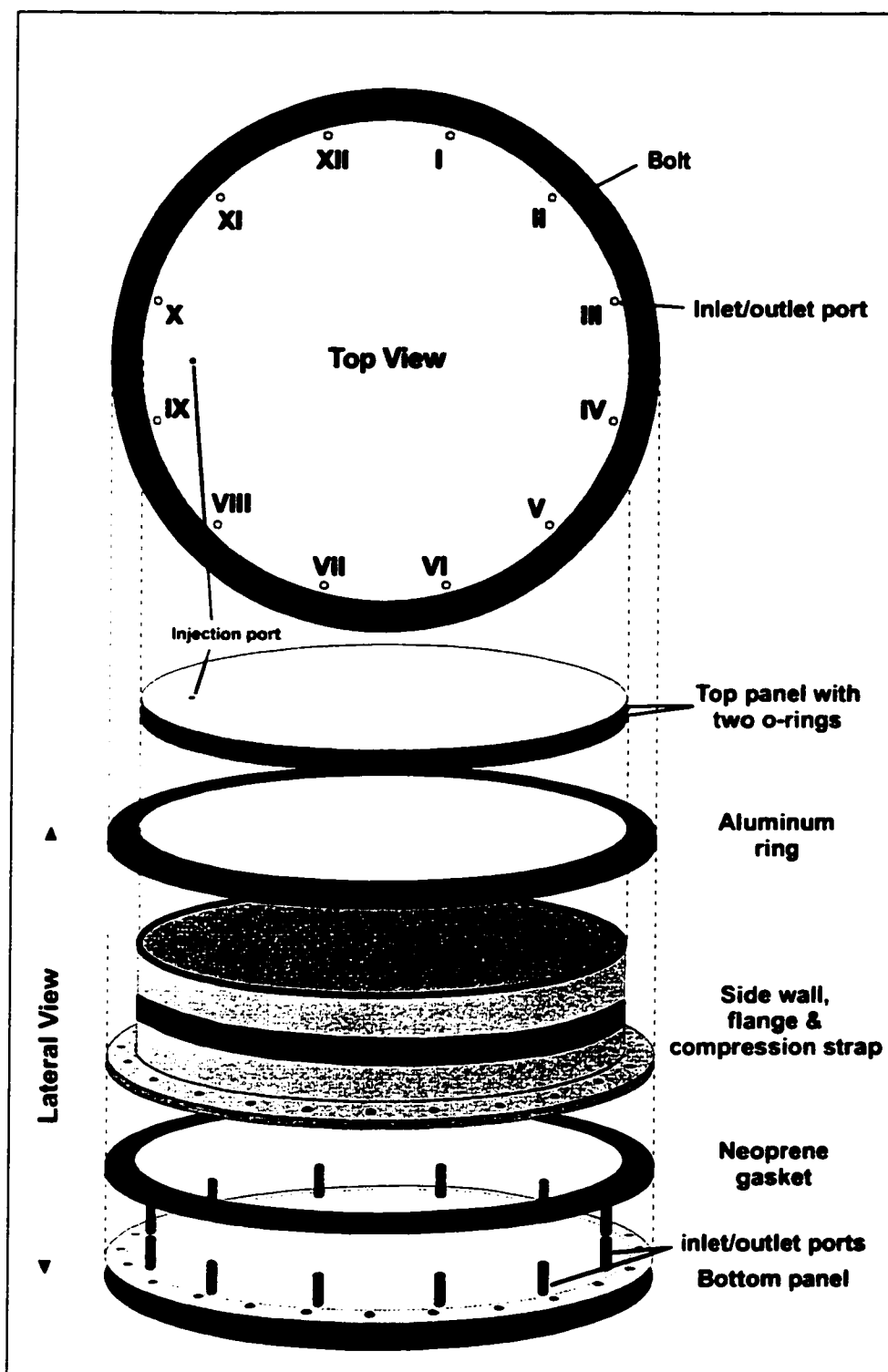


Figure A-2. Schematic diagram of experimental flow cell (not to scale). See text for a detailed description of the individual components.

The cover was secured in place, tightly pressed against the porous medium using 6 adjustable clamps, which were kept in place to prevent the cover from shifting, as shown in Figure A-3.

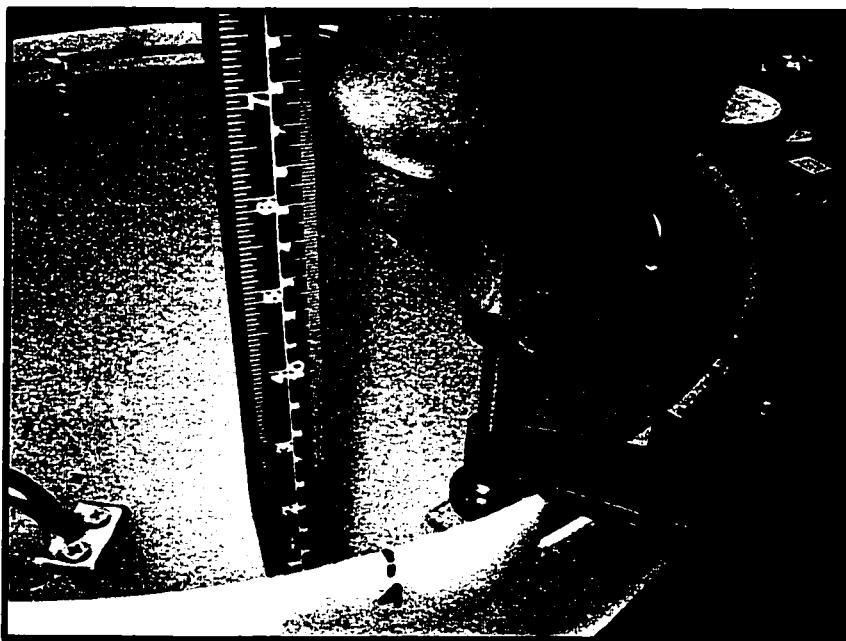


Figure A-3. Image showing the insertion of the flow cell cover. The Plexiglas[®] cover was machined to fit precisely inside the cell's lateral wall, and fitted along its circumference with a soft rubber band (weather stripping material) between a pair of two o-rings to provide a watertight seal with the cell wall. Six adjustable clamps (only one shown here) were used to lower the cover evenly until its bottom surface lay flat against the top of the water-saturated porous medium. The clamps were left in place to prevent the cover from shifting.

Preliminary experiments revealed that the two o-rings were not sufficient to maintain a leak-free seal between the hard Plexiglas[®] cover and softer PP wall. A strip of commercially available spongy weather stripping (normally used as a seal around doors and windows) was used between the two o-rings to palliate the leakage problem. The seal was improved further using a ratcheted canvas belt placed around the circumference of the lateral wall (see Figure A-4).

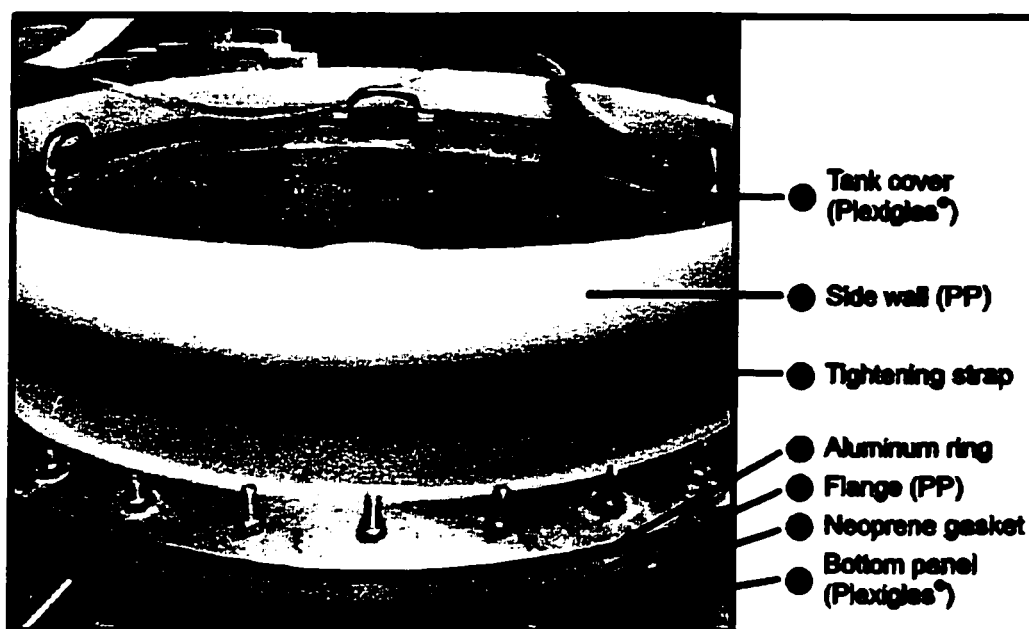


Figure A-4. Annotated image showing a lateral view of the experimental flow cell shown with emplaced cover. Note the canvas strap wrapped tightly around the walls, that was used to improve seal between the cell cover and sidewalls.

1.2 Hydraulic control system (HCS)

The hydraulic control system (HCS) is possibly the most critical part of the experimental setup, since precise regulation of the hydraulic heads at each of the 12 inflow/outflow port locations is essential to produce a uniform flow field. The main objective of this study – to determine the impact transient flow on solute dispersion – necessitated the design of an accurate flow control system that allowed rapid changes to the flow direction and/or magnitude. The HCS was comprised of four principal components: (1) the hydraulic ports inside the flow cell, (2) the multi-channel hydraulic switch, (3) the input/output reservoirs, and (4) the overflow manifold. The four components were connected using clear flexible Tygon[®] tubing (internal diameter 0.635 cm), as illustrated in Figure A-1.

Water flows into the model from input reservoirs, through the hydraulic switch and enters the porous medium via inlet ports. Water exits the model through outlet ports and makes its way to the outlet reservoirs via the hydraulic switch. The hydraulic switch determines which of the 12 ports will act as inlet ports, and which will act as outlet ports. The hydraulic head

in the input/output reservoirs is determined by the position of the inlet/outlet overflow on the inlet/outlet manifold. Figure A-5 shows a simplified view of the HCS, illustrating the pathway travelled by a hypothetical water molecule that flow into and out of the system. For the sake of simplicity, only two channels are shown. A detailed description of each of the HCS component follows.

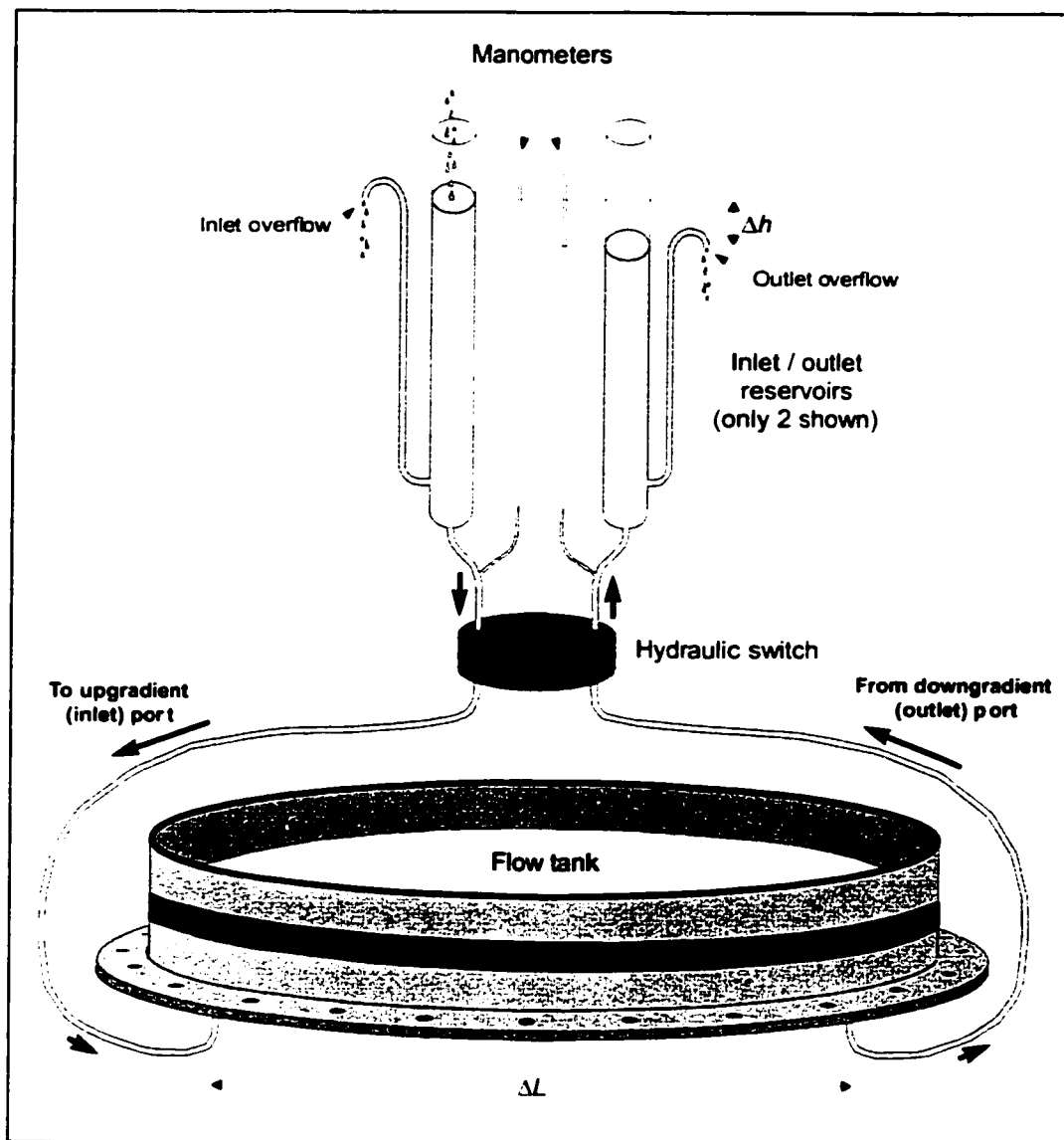


Figure A-5. Schematic representation of the hydraulic control system (HCS), showing the path followed by the water as it travels from the upgradient (inlet) reservoirs, through the flow cell (packed with a porous medium and saturated), and exits the system at the downgradient (outlet) reservoirs. Each of the cell's 12 hydraulic ports is connected to a separate reservoir via the multi-channel hydraulic switch. The switch allows instantaneous variations in mean flow direction inside the cell. The bulk hydraulic gradient is defined as $\Delta h / \Delta L$, and can be controlled by adjusting the water levels in the reservoirs.

1.3 Hydraulic ports

Twelve inlet/outlet hydraulic ports were used to establish the desired hydraulic head distribution inside the flow cell. The 12 ports were located inside the flow domain, 8.25 mm away from the cell wall, at positions along the circumference that correspond to the hours positions on a clock, resulting in a 30° radial angle between adjacent ports (see Figure A-2). Each port was made of copper tubing, 2.75 cm high, with an internal diameter of 0.635 cm. The tubing was slotted longitudinally from top to bottom at three equally spaced locations around the circumference, producing three “windows”, which covered approximately 62% of the internal surface area. The windows were screened using a 170 µm stainless steel mesh, providing efficient hydraulic contact with the surrounding porous medium. Each port was closed at the top and connected at the bottom to one of the input/output reservoirs via the hydraulic switch (see below).

1.4 Multi-channel hydraulic switch

The main purpose of the hydraulic switch (see Figure A-1(b) and Figure A-5) is to allow the average groundwater flow direction to be modified almost instantaneously, without interrupting the experiment. The switch couples each of the inlet/outlet ports to an inlet/outlet reservoir of prescribed hydraulic head; the hydraulic head distribution in the flow cell can thus be changed nearly instantaneously by rotating the switch.

The switch is composed of two solid acrylic disks (each 15.24 cm in diameter and 2.54 cm thick), held together in their centre to allow them to be rotated against each other. The disks have a matching set of 12 openings, each 0.625 cm in diameter and each equipped with a fitting for flexible tubing. The 12 fittings on one plate are connected to the 12 hydraulic ports in the model; and the 12 fittings on the other plate are connected to the 12 input/output reservoirs. All tubing connections are made of 0.635 cm (1/4”) I.D. Tygon® tubing. The holes in the plates are lined up by means of a spring-mounted 0.953 cm (3/8”) ball bearing in one plate with matching circular depression in the other plate. Neoprene o-rings (0.32 cm diameter) between the plates and around each of the 12 holes maintain a hydraulic seal between the plates for each set of lined up holes. The pressure exerted on the

o-rings is set by the tightness of the two plates against each other, which can be adjusted by means of a nut and bolt and spring assembly at the centre of the plates.

To rotate the bulk groundwater flow direction, one disk is rotated with respect to the other, thereby connecting the hydraulic ports to reservoirs of different preset hydraulic head. For example, to rotate the average flow direction by 30°, the disk is rotated by one step clockwise or counter-clockwise. This device had the double advantage of allowing quasi-instantaneous direction shifts in flow direction, and doing so without disturbing the static hydraulic heads in the input/output reservoirs (see next section). Additionally, it was possible to temporarily stop flow inside the flow cell, by rotating one of the disks only halfway between flow through positions. This feature is quite useful, for example, during tracer injection to minimize smearing of the initial plume.

1.5 Input/output reservoirs

The only means for water to flow in or out of the experimental model is through the inlet/outlet ports described previously. Upgradient (input) ports supply water to the cell at the same rate that it is removed by the down gradient (output) ports. The mass of water flowing into or out of the cell at each individual port is controlled by the relative magnitude of the hydraulic heads imposed at each port location. The hydraulic head was regulated by a system of 12 reservoirs connected to the 12 ports of the flow cell via the hydraulic switch, as shown in Figure A-1(a) and (c). The hydraulic head in each reservoir was maintained at a specified level by overfilling the reservoir with a constant source of water and allowing the overflow to drip from a specified level through the overflow manifold. This produced an constant water supply in each reservoir, so that the corresponding port acted as a “specified head boundary”, either adding or removing water from the system as imposed by the flow conditions at that location [Anderson and Woessner, 1992]. The hydraulic head was monitored with a manometer connected to each reservoir.

1.6 Overflow manifold

Key to establishing the desired flow configuration in the system was the precise regulation of the hydraulic head in each channel. For this purpose, 12 pipette tips were installed along the perimeter of a circular disk at regular radial intervals, thus replicating the clock-like geometry of the input ports inside the flow cell (see Figure A-1(d)). The angular separation between adjacent regulators was exactly 30° , the same as the hydraulic ports in the flow cell. The elevation of each tip on the disk controlled the water elevation (pressure head) in the corresponding water supply reservoir. Tilting the disk to a desired angle thus controlled the magnitude of the hydraulic gradient inside the flow cell. The mean hydraulic gradient inside the cell is obtained from $i = \Delta h / L$, where Δh is the head difference between the upstream and downstream channels, for the ports separated by a distance L . Hydraulic heads were monitored individually for each channel using manometers, located between the supply reservoirs and the flow cell.

2 POROUS MEDIA

The porous media used throughout the experiments were composed of glass oxide beads of various sizes (Potters Industries¹). These beads were selected because of their spherical shape, relatively uniform size distribution within a given class, high translucence, and low affinity for sorption with the tracer used. Section 3.1 presents a summary of a laboratory batch test conducted to determine the distribution coefficient.

¹ Potters Industries Ltd, 1505 Industrial Blvd, La Prairie, Quebec, Canada, J5R 2E4, (450) 659-9129

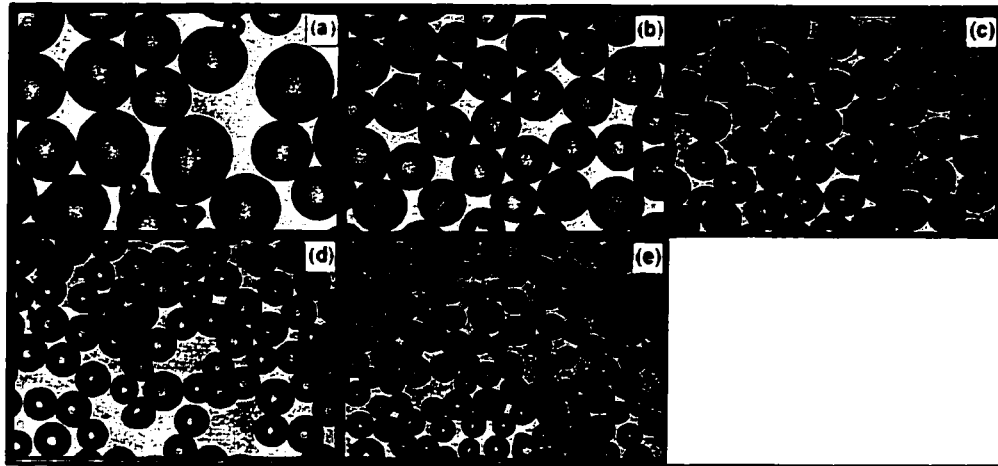


Figure A-6. Photographs of the glass beads taken under the microscope for bead classes B (a), C (b), AA (c), AB (d) and AC (e). Field of view for each picture is approximately 2.5 cm (magnification x32).

2.1 Physical properties

The physical properties of the various classes of porous media used are reported in Table A-1. Laboratory measurements of hydraulic conductivity were made as part of a parallel study [Pion, 2001].

Nine distinct classes were used, four of which were obtained by thoroughly mixing an equal volume of beads from two adjacent size classes. For example, class 2 (B+C) was produced by combining beads B and C with a 1:1 ratio.

Table A-1. Physical properties of porous media

^{a,b} Bead class	ASTM sieve nos.	^c Diameter (μm)	Porosity	^e Hydraulic conductivity (cm s ⁻¹)	^f Standard deviation (cm s ⁻¹)	^g Kozeny-Carman (cm s ⁻¹)
1 (B)	30/40	420-595 (508)	0.38 (<i>n</i> = 2)	0.182	6.9 x 10 ⁻⁴	0.200
2 (B+C)	30/60	250-595 (423)	0.38 (<i>n</i> = 2)	0.124	2.0 x 10 ⁻³	0.139
3 (C)	40/60	250-420 (335)		0.105	5.6 x 10 ⁻⁴	0.087
4 (C+AA)	40/70	210-420 (315)	0.38 (<i>n</i> = 2)	0.078	8.6 x 10 ⁻⁴	0.077
5 (AA)	40/70	210-420 (315)		0.072	6.7 x 10 ⁻⁴	0.077
6 (AA+AB)	40/80	177-420 (299)	0.38 (<i>n</i> = 2)	0.053	3.1 x 10 ⁻⁴	0.069
7 (AB)	50/80	177-297 (237)		0.045	6.9 x 10 ⁻⁴	0.044
8 (AB+AC)	50/100	149-297 (223)	0.38 (<i>n</i> = 2)	0.030	3.4 x 10 ⁻⁴	0.039
9 (AC)	60/100	149-250 (200)		0.026	9.1 x 10 ⁻⁵	0.031

b. Manufacturer bead class shown in brackets

c. Mixed classes (nos. 2, 4, 6 and 8) produced from adjacent classes using a 1:1 ratio

d. Range corresponding to ASTM sieve openings (arithmetic mean in brackets)

e. From *Bear* [1972], eqn. 5.10.18. (see text for discussion)

f. From permeameter measurements using several flow rates (all numbers are averages of 12 measurements)

g. Standard deviation of 12 measurements

Porosity

Porosity (*n*) is defined as the ratio of the volume of voids (V_v) to the bulk (total) volume (V_b) of a porous medium. For unconsolidated porous media such as the glass beads used here, porosity is dependent upon the packing, shape, arrangement and particle size distribution [*Bear*, 1972].

Duplicate porosity measurements were made by filling a 100 mL graduated cylinder with selected classes (listed in Table A-1), and recording the dry weight W of the 100 cm³ samples. Bulk density was obtained from $\rho_b = W / V_b$, and used to estimate porosity from the following relationship [*Freeze and Cherry*, 1979]:

$$n = 1 - \frac{\rho_b}{\rho_s} \quad (\text{A.1})$$

where $\rho_s = 2.50 \text{ g cm}^{-3}$ is the particle mass density of the silica beads, as reported by the manufacturer.

The porosity of uniformly packed spheres of equal diameters can vary from 0.26 for rhombohedral packing arrangement, to 0.48 for cubic packing, regardless of sphere size [*Benenati and Brosilow*, 1962]. Porosity was measured experimentally for bead classes 1, 2, 4, 6 and 8, and was found to be equal to 0.38. This compares well with results reported by

Schincariol et al. [1993], who used beads from the same manufacturer in their experiments. A porosity value of 0.38 was therefore used throughout this study for all bead classes.

Permeameter measurements

The hydraulic conductivity of each bead class was measured using a constant head permeameter. The permeameter was constructed by adapting the design of *Driscoll* [1986] (Figure 5.13, p. 74). It consists of a transparent Plexiglas® cylinder (8.25 cm internal diameter × 20 cm height) fitted at each extremity with end caps that hold the porous medium sample being tested inside the cylinder. Each end cap is fitted with a tubing adapter and a porous stone that allows water into the porous medium at one end of the cylinder, and out at the other end. The porous plate prevents the loose granular materials packed inside the permeameter from clogging the tubes, and produces a uniform pressure across the face of the porous medium. The method used to determine permeability was: “Standard test method for permeability of granular soils (constant head)”, American Society for Testing and Materials (ASTM) standard D2434-68(1994)e1. A summary of the test procedures is presented here.

After inserting a porous stone disk at the bottom of the permeameter, distilled water was added to a height of a few centimetres. The sample was then loaded by pouring a small amount of the silica beads at a time, using increments of 2-3 cm, while adjusting the water level to maintain saturation of the sample at all times during loading. To ensure optimal packing density, each layer was thoroughly mixed using a rod and vibrated by tapping the sides of the permeameter with a rubber mallet and compacting the sample using a piston-like plate before adding the next layer. Upon completion, the column was compacted by pressing the adjustable top plate, equipped with a second porous disk, on top of the sample.

After sealing the permeameter, the outlet valve was adjusted to establish a constant hydraulic gradient within the cell. After steady conditions were reached (approximately 5 min), a graduated cylinder was placed to collect water at the outlet, and water levels h_1 and h_2 in the piezometers were recorded.

Water levels (hydraulic heads) were measured in two manometers (h_1 and h_2 , respectively) connected to pressure ports at two locations separated by a distance $L = 9.25$ cm in the longitudinal direction (parallel to flow). The flow rate Q was estimated by measuring the total volume of water V_w collected at the outlet in a graduated cylinder in a given amount of time t . Darcy's Law [e.g. *Bear*, 1972] was then used to derive the sample's hydraulic conductivity K :

$$K = \frac{Q}{A} \frac{(h_1 - h_2)}{L} \quad (\text{A.2})$$

where $Q = V_w / t$, A is the sample's cross sectional area (perpendicular to the direction of flow), and $(h_1 - h_2) / L$ is the hydraulic gradient through the sample.

A nested experimental design was used to determine the sources of variability in the method [*Pion*, 2001]. For each bead class, two samples (packings) were analyzed; for each packing, three flow rates were used; and at each flow rate, two replicate measurements were made. Each measurement consisted of a flow rate reading and of a pair of manometer readings. There were therefore 12 measurements for each bead class (2 packings $\times$ 3 flow rates $\times$ 2 replicates). The geometric mean of all measurements and associated standard deviations are shown in Table A-1.

Permeability k was also estimated using the Kozeny-Carman equation [*Bear*, 1972]:

$$k = \frac{[n^3 (1-n)^2]}{5M_S^2} \quad (\text{A.3})$$

where M_S is the specific surface of the porous media, defined as the "total interstitial surface area of the pores per unit bulk volume of the porous medium" [*Bear*, 1972 (p. 50)]. Porosity n was 0.38 for all bead classes. Specific surface was estimated using $M_S = 3 / r$, using the mean grain diameter r for each class. Hydraulic conductivity values derived from this equation and reported in Table A-1 were calculated using [*Freeze and Cherry*, 1979]:

$$K = 9.80 \times 10^4 k \quad (\text{A.4})$$

where k is expressed in cm^2 and K is in cm s^{-1} .

As can be seen in Table A-1, the agreement was excellent between the permeameter measurements of hydraulic conductivity and those estimated from the Kozeny-Carman equation. This is especially true when considering the empirical nature of the Kozeny-Carman relation. This good agreement is perhaps because the glass beads are close to what can be considered an “ideal” porous medium, being both spherical and well-sorted (see Figure A-6).

2.2 Packing method

As stated by *Bear* [1972], obtaining a uniform saturated packing, where the characteristics of the porous medium are uniform and air is completely excluded, is a difficult task. In traditional thin-slab (pseudo two-dimensional) flow cells, porous media are added through an opening at the top of the cell [e.g. *Schincariol*, 1989; *Glass et al.*, 1995; *Norton and Glass*, 1993]. Differential settling due to gravity often results in some layering – and preferential flow pathways – that may greatly influence the migration of dissolved solutes. In the case of heterogeneous porous media, geometry is generally limited to simple cases of layered systems due to technical difficulties associated with packing the flow cell from the top. To address these issues, an original design was developed that allowed porous media to be added from the front of the flow cell. This was made possible by the removable front cover of the flow cell.

Emplacement of the porous medium (“loading”) was a critical part of the experiments, and particular attention was given to this crucial step. The primary consideration during loading was to ensure that the transmissivity, T (hydraulic conductivity times the thickness), was uniform for a given grain size. This meant that T had to be constant across the entire flow cell when building a homogeneous porous medium, and locally for each conductivity class when heterogeneous media were constructed. In the case of heterogeneous porous media there was the added complexity of spatially varying conductivity in a puzzle-like arrangement consisting of a number of zones (or cells) of desired dimensions, each filled with a single bead class of homogeneous properties.

Practically, the requirements of uniform packing in the z dimension (thickness) and uniform transmissivity in the x - y (horizontal) plane for a given bead class meant that beads had to be put in place with the following considerations in mind:

- ◆ Minimal layering (and thus hydraulic conductivity variations), caused by gravitational settling due to bead size variations in the z dimension;
- ◆ Uniform bead thickness in the z dimension;
- ◆ Uniform packing density across the cell; and
- ◆ Ability to create heterogeneous porous media according to some pre-determined x - y distribution (deterministic or stochastic).

The front-loading model design used here is unique and offers several advantages over the traditional procedure that consists in loading the sand – or beads – into the flow model from the top edge. These advantages include:

- ◆ Porous medium can easily be mixed (homogenized) to remove any layering created during emplacement, either across the entire cell for homogeneous experiments or within each zone in the case of heterogeneous media;
- ◆ Thickness of the flow domain, while uniform for the entire model, can be easily adjusted by adding the appropriate volume of granular materials (the cover's position will vary accordingly);
- ◆ Packing density variations are reduced by pressing the cover onto the porous medium; and
- ◆ Heterogeneous porous media can be more easily constructed by using templates shaped and arranged to create the various zones where beads of contrasting permeability can be packed (templates are gently removed prior to cover emplacement).

Homogeneous media

To minimize layering in the porous medium, the beads were poured into the open cell while maintaining the water level a few centimetres above the beads, thoroughly mixed (by hand) and carefully levelled to a final thickness of approximately 33 mm, sufficient to cover the inlet/outlet ports by a few mm. Any unintentional layering introduced during bead emplacement was destroyed by the mixing process, as were entrapped air bubbles. Although no porous medium is truly homogeneous, the front-loading method resulted in porous media virtually free of layering, in any direction.

The flow cell cover was then positioned on top of the supernatant water, with a small angle to allow the escape of all air bubbles caught between the water surface and cover. The cover was then carefully lowered using six threaded C-clamps, until its bottom surface and the top of the porous medium were in contact across the cell. Water pressure created by the insertion of the cover was released by allowing water to escape through the I/O ports. This process had to be gradual to avoid extreme flow rates out of the ports that could disturb the porous medium in their vicinity. The clamps were then gradually tightened on alternating sides of the flow cell to ensure that the thickness of the flow domain remained uniform, and that the porous medium was evenly packed. As a final precaution to eliminate any void space at the interface between the porous medium and the flow cell wall, the flow cell was placed on edge in a vertical (upright) position, and vibrated using a rubber mallet. The absence of a void space at the wall surface near the top of the cell was an indication that packing was successful. Had the cell not been properly packed, the porous medium would have settled when the cell was vibrated, and an empty space created at the top wall. The cell was then placed in front of the light box and left in its vertical position throughout the duration of the flow experiments.

Heterogeneous (stochastic) media

Because of the front-loading cell design, we were successful in constructing heterogeneous porous media of specified statistical properties. For this purpose, a physical template was used to subdivide the flow domain into small compartments. The template consisted of thin-

walled plastic light diffusers used in fluorescent lighting that were glued together to produce the required thickness, and machined to a circular shape to fit precisely inside the flow cell. The template divided the open flow cell into approximately 1450 compartments, each 1.5×1.5 cm in the horizontal plane and 5 cm high. The volume occupied by the template walls was taken into account when measuring the volume of glass beads required within each compartment to produce the target final thickness, sufficient to cover the I/O ports completely once the template was removed. Each compartment of the template was filled with one of the nine classes of glass beads presented in Table A-1. The arrangement was determined by sampling a random distribution of correlated hydraulic conductivity values derived stochastically. This method is an adaptation of a technique developed for one-dimensional porous media by *Welty and Elsner* [1997].

Random field generation

Two-dimensional random fields were generated numerically using the technique of *Robin et al.* [1993]², where the average hydraulic conductivity, variance and correlation scales were specified as input parameters. Although their full dimensions were set to 1024 by 1024 cells², the random fields were truncated to 700 by 700 cells² to avoid banding problems associated with periodicity inherent to the technique. The nodal spacing was chosen to be 0.151 cm in both horizontal dimensions (x and y), and the truncated field dimensions were thus 105.8 x 105.8 cm². The mean and variance for $\ln(K)$ used as input to the random field generator were -2.74 and 0.40, respectively. These values were selected to approximately match the mean (-2.71) and variance (0.42) for the log hydraulic conductivity that were measured in the laboratory for the bead classes, as discussed in § 2.1. The statistical model used in the random field generation was an exponential decay autocovariance function with correlation scales (decay constants) of 6.0 cm in both x and y directions [c.f. *Robin et al.*, 1993] to produce an isotropic field.

² For information about the random field generator *FGEN*, contact the author, Dr. Michel J.L. Robin, University of Ottawa, Department of Earth Sciences, Ottawa, Ontario, Canada.

Random field sampling

Because the numerical random fields had a relatively fine grid spacing ($\Delta x = \Delta y = 0.151$ cm), they had to be sampled in order to assign the appropriate bead class to each of the larger blocks (1.51 cm by 1.51 cm) in the flow cell. This was done by first extracting the nodal hydraulic conductivity values from a 65 cm by 65 cm zone located near the centre of the random field. A new field was then produced with *FGEN* 9.2 [Robin *et al.*, 1993], with the same overall dimensions but a coarser grid that matched the desired compartment size. Each block on the new grid was assigned a hydraulic conductivity value obtained from the geometric mean of the 100 smaller blocks on the original grid. The resulting random fields have the prescribed correlation structure except at scales smaller than 1.51 cm, where the autocorrelation is forced to a value of 1.0. The mean and variance for the sampled grid (43 blocks by 43 blocks) were computed and compared against the same statistics for the original grid, to ensure they were similar. If the pre- and post-sampling values were significantly different, a new random field was generated – using a different seed – and then sampled, until an acceptable field was produced. Figure A-7 illustrates the sampling procedure.

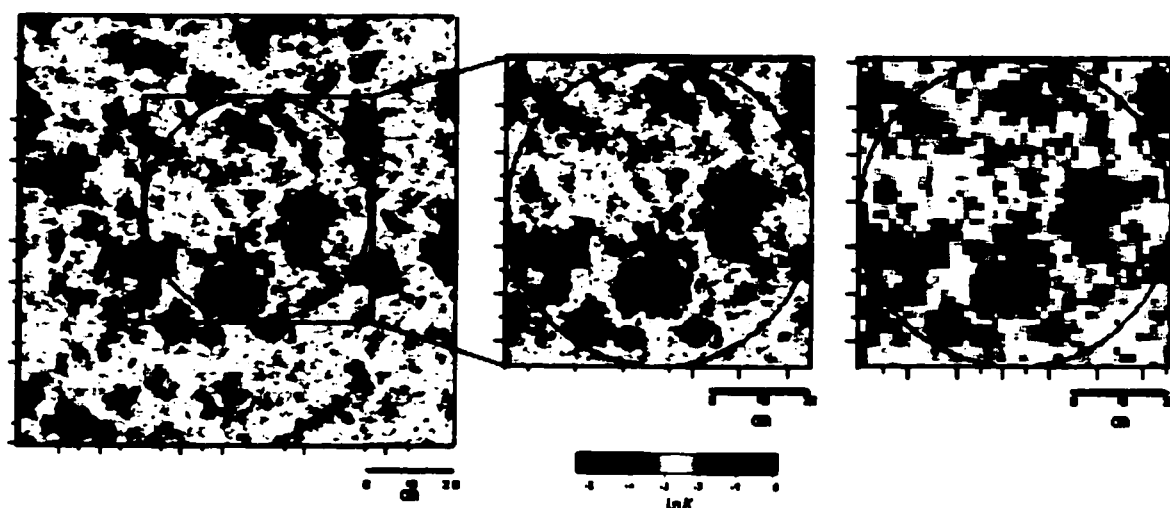


Figure A-7. Random field sampling procedure. A subset (b) of the generated field (a) is extracted, and sampled using a coarser grid and grouped into bins to generate the final field (c) used to map the hydraulic conductivity values when loading the flow cell with the heterogeneous porous medium.

The final step was to assign to each block the hydraulic conductivity values obtained from the permeameter measurements for each bead class. The sampled numerical data were thus sorted into nine bins with bounds defined by the geometric mean between two adjacent bead classes, as shown in Table A-1. Each compartment of the template was populated with a measured volume of class beads, using the map binned bead classes shown in Table A-1.

Table A-2. Beads size classes used in Table A-1. $\ln K$ ranges for each class are from Figure A-7. K was estimated experimentally using a constant head permeameter setup (see Appendix A for further details).

	Beads	In K range		Bin	count	K cm/s
		from	to			
1	B	-1.9	-1.72	228	0.182	
2	B+C	-1.9	-2.28	229	0.124	
3	C	-2.28	-2.51	185	0.105	
4	C+AA	-2.51	-2.59	46	0.078	
	AA	-2.59	-2.79	157	0.072	
6	AA+AB	-2.79	-3.02	160	0.053	
	AB	-3.02	-3.3	160	0.045	
8	AB+AC	-3.3	-3.58	115	0.030	
9	AC	-3.58	-3.67	148	0.026	

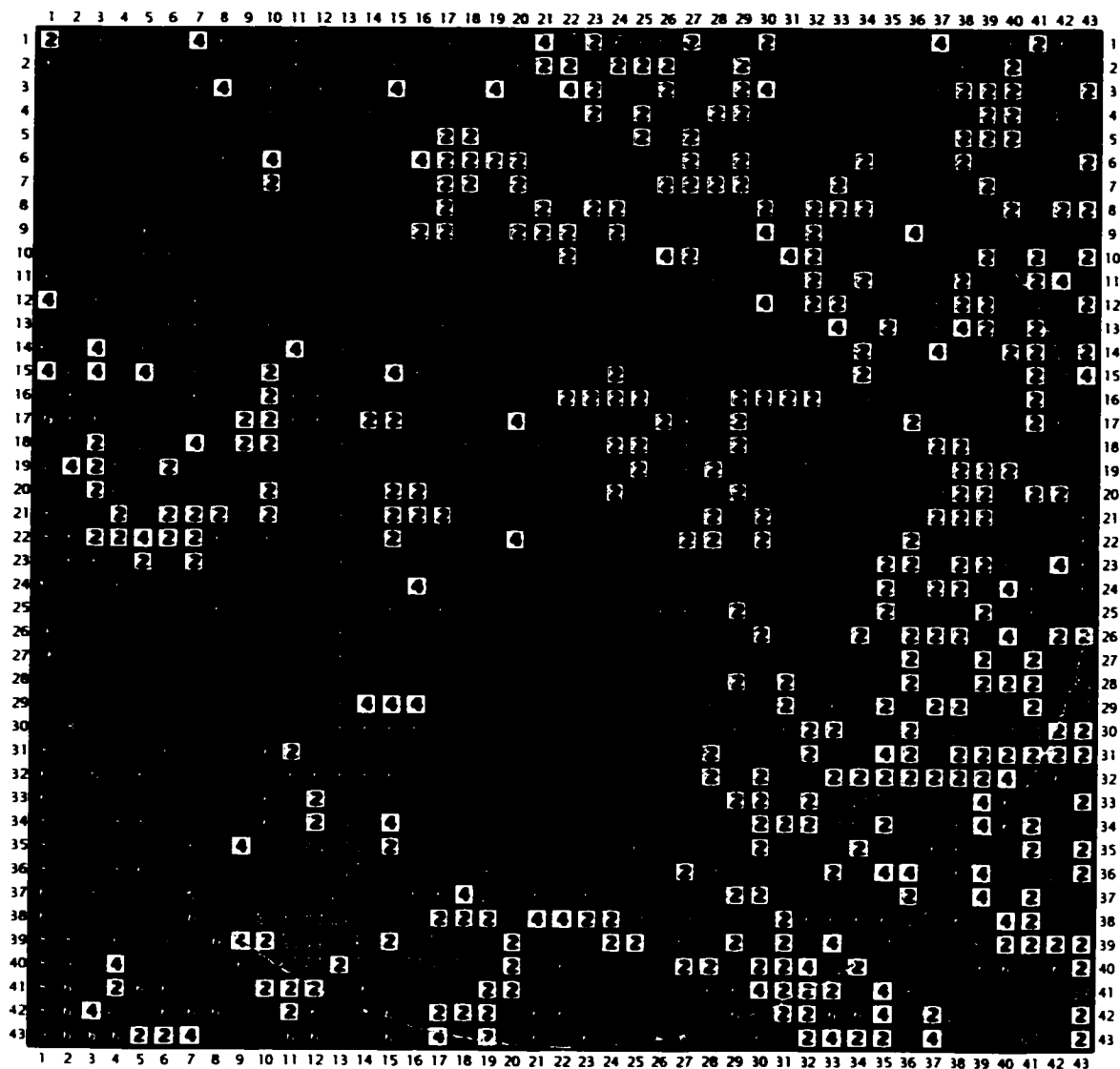


Figure A-8. Map of the log-normally distributed hydraulic conductivity values ($\ln[K]$) used to construct the heterogeneous porous medium. This distribution was randomly generated on a finer grid (430×430), and then reduced to 43×43 by averaging 100 $\ln[K]$ values in each cell (see Appendix A for details about the random field generation). Each block was assigned a bead class (numbered 1-9) based on its averaged $\ln[K]$ according to Table A-1. The outline of the circular flow domain is shown in yellow.

Means and variances for the “binned” fields were also calculated after replacing each cell with the hydraulic conductivity value determined by the bins from Table A-2, and compared with the statistics from the original (700 by 700), sampled, and binned fields (both 43 by 43), as shown in Table A-3.

In general, the sampling and “binning” had little impact on the means. Variance, in contrast, was inevitably reduced by replacing block hydraulic conductivity values by their assigned bin values.

Table A-3. Comparative statistics of random field generated using *FGEN* 9.2, using 111 as seed number.

	Mean $\ln K$	σ^2
Input (target values)	-2.74	0.4
Generated field (700x700)	-2.67	0.45
Sampled field (43x43)	-2.66	0.52
Binned field (43x43)	-2.62	0.37

Flow cell loading

The map containing the bead class distribution was printed to scale and placed under the flow cell to serve as a guide. Figure A-8 shows a reduced version of the map, where the codes (1-9) correspond to the hydraulic conductivity classes defined by the property numbers listed in Table A-1. Each bin was colour coded to make the filling process easier. Distilled water was added to a height of a few centimetres, and the plastic grid used to partition the individual cells was inserted into the cell.

Approximately 1450 cells were filled one by one with the glass beads corresponding to the codes on the map. Figure A-9 shows a summary of the emplacement procedure used to construct the heterogeneous porous medium. Template compartments located in the area surrounding each of the 12 inlet/outlet ports were cut out to make room for the ports. These zones were filled with high permeability Type 3 beads ($K = 0.105 \text{ cm s}^{-1}$). The cells located along the perimeter of the cell were partially truncated to fit the circular geometry, and were filled with low permeability Type 9 beads ($K = 0.026 \text{ cm s}^{-1}$). Finally, after loading all compartments, a thin layer (approximately 1 mm) of Type 9 beads was spread across the entire flow cell, in order to compensate for the slight thickness variations from cell to cell. This top layer was also intended to minimize wall effects caused by forcing spherical grains against the flat cover. Because wall effects are directly related to grain diameter [e.g. *Benenati and Brosilow*, 1962], their relative importance is expected to be greater for large

diameter beads than for finer grains. Since bead diameter varies spatially for heterogeneous media, adding a thin uniform layer over the other types of beads palliated this problem.

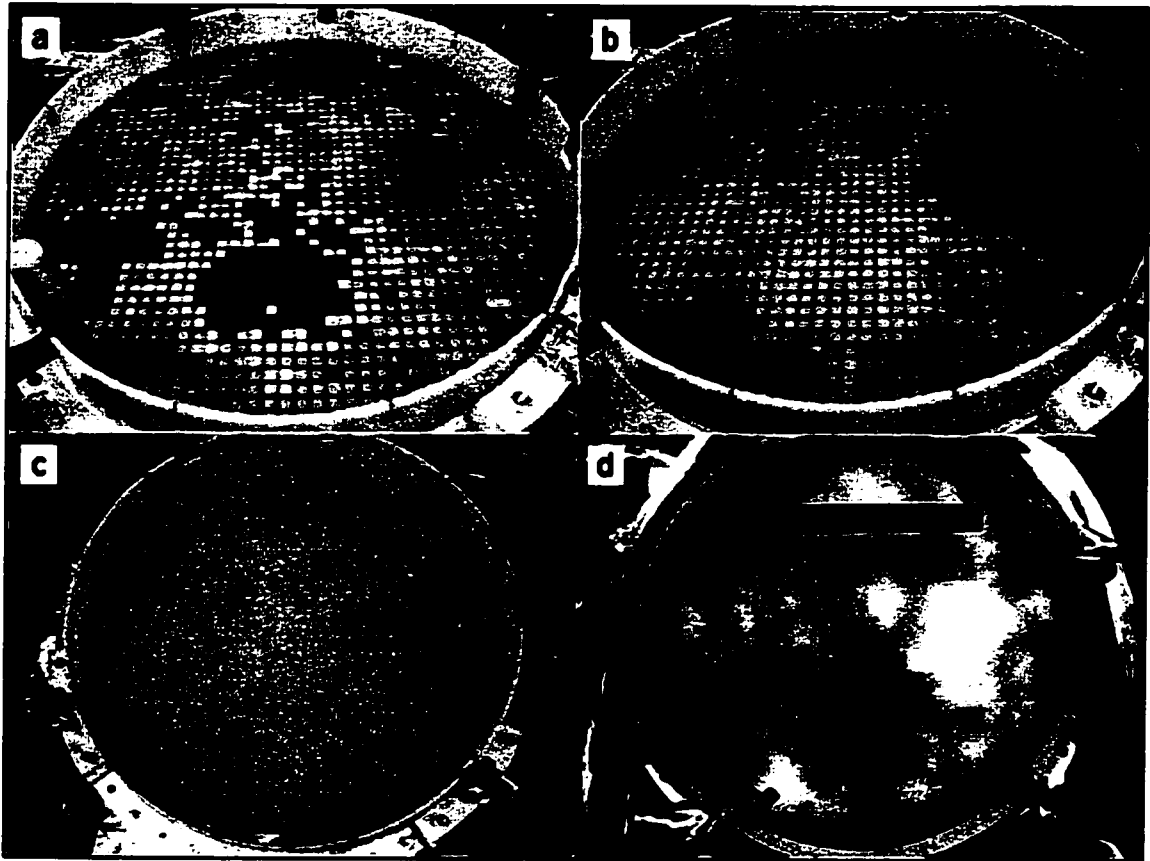


Figure A-9. Summary of the emplacement procedure used to construct the heterogeneous porous medium. Each of the template's $1.5 \times 1.5 \text{ cm}^2$ blocks (about 1450 in total) was filled with a pre-measured volume of glass beads according to the map shown in Figure A-8. The image in (a) shows the flow cell after bead classes 1-7 had been added. Once all complete blocks (those not partially truncated by the circular outline of the flow cell) were filled (b), the finest beads (class 9) were used to fill the partial blocks around the flow domain. The template was then gently removed (c), and the clear Plexiglas® cover firmly inserted to seal the water-saturated flow cell. The entire assembly was placed upright in front of the light box (d). Note that the entire procedure shown here took place under water to avoid air entrapment within the porous medium.

Finally, the cover was emplaced and secured using clamps, and the cell vibrated and placed vertically in front of the light box, as described previously for homogeneous media.

3 TRACER

Following the rationale of *Schincariol et al.* [1993], the tracer selected for this study was Intracid[®] Rhodamine WT (RWT). This tracer is a bright fluorescent red dye manufactured specifically for water-tracing by Crompton and Knowles³. Its high tinctorial strength, low affinity for sorption, chemical stability and high solubility were among the factors influencing this selection. RWT was selected also because it poses no significant environmental or health risks [Douglas, 1983]. Finally, the high sensitivity of solid-state digital cameras (often referred to as charge coupled devices, or CCD) to the red portion of the visible spectrum [Russ, 1995] was also taken into consideration in selecting this tracer.

RWT is available as a concentrated liquid solution from the manufacturer. The exact physical properties may vary slightly depending on the manufacturing lot. Consequently, a number of physical properties were measured specifically for the RWT batch used in this study, namely density, concentration and sorption coefficient. The details of these measurements are given in this section.

3.1 Tracer properties

Physical properties

A compromise had to be reached in determining the most appropriate dye concentration to use. To enhance concentration contrasts, the colour intensity of the dissolved tracer had to be as high as possible, while making sure the density of this solution was small enough so that gravity-induced effects were negligible. As a first step it was essential to know the exact concentration and density of the stock solution.

Two ceramic beakers were oven-dried for approximately one hour and allowed to cool to room temperature. The beakers were weighed empty, and 3.00 mL of de-ionized water was added to each. A 1.00 mL aliquot of the concentrated RWT solution was then added and the beakers were weighed once more to determine the density of the resulting solutions. Finally, the solutions were allowed to evaporate in an oven for approximately 100 hours and the dry

³ Crompton and Knowles Colors Incorporated, P.O. Box 33188, Charlotte, NC 28233, (704) 372-5890

residue was weighed to calculate the solution's concentration. Results are summarized in Table A-4.

Table A-4. Physical properties for concentrated Rhodamine WT averaged from the two experimental vessels, and corrected at 20°C.

Property	Average	Standard deviation, s
Density, ρ (g cm ⁻³)	1.0862 ($n = 2$)	0.0011
Concentration, C (g L ⁻¹)	357.7 ($n = 2$)	0.4243

A large volume (about 6 L) of RWT stock solution was prepared and used throughout this work. This stock solution was prepared by diluting 2.7618 mL of the manufacturer's concentrate into 6.0 L, producing a concentration of $C_{stock} = 164.7 \text{ mg L}^{-1}$. The stock solution was diluted using distilled water to produce solutions of desired concentration C_0 , to be injected into the flow cell as a tracer. Concentration values were obtained by measuring the absorbance of the solutions using a spectrophotometer and deriving concentration values from a calibration curve. Readings were made by setting the instrument to a fixed wavelength ($\lambda = 556 \text{ nm}$) corresponding to the optimal absorption wavelength for RWT. The calibration curve, giving the relationship between C and A , was measured using RWT solutions of known concentration. The linear regression ($R^2 = 0.9979$) presented in Figure A-10 was then used to determine a solution's concentration from its absorbance:

$$C = A / 0.124 \quad (\text{A.5})$$

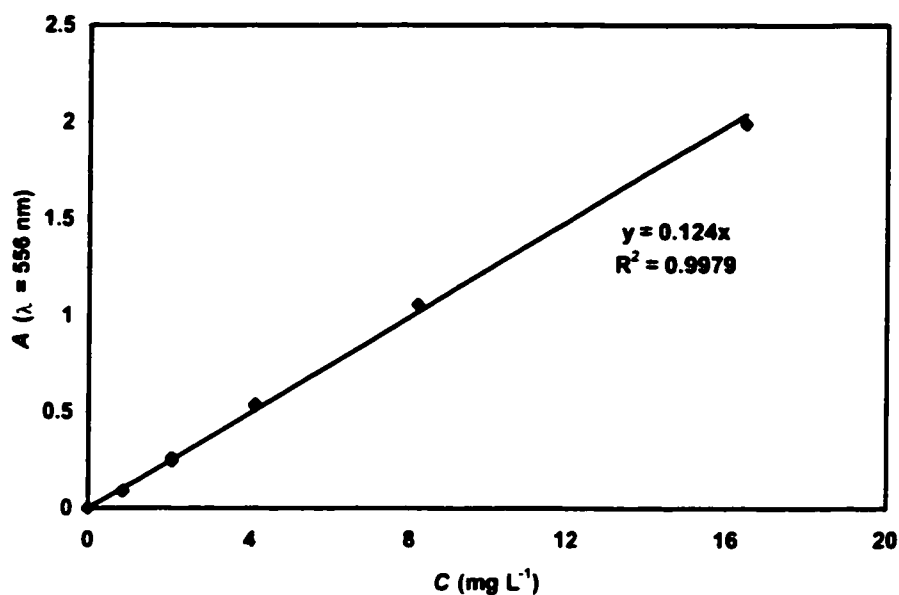


Figure A-10. Rhodamine WT (RWT) calibration curve. Absorbance A measured at $\lambda = 556$ nm, which corresponds to the tracer's optimal absorption wavelength.

Distribution coefficient (K_d)

A laboratory batch experiment was conducted to determine the distribution coefficient, K_d , for RWT on the silica beads. K_d is a direct measure of the partitioning between the solid (sorbed) and aqueous (dissolved) phases of a contaminant [U.S. Environmental Protection Agency, 1999a]. It can be described as the concentration of tracer attached to the solid (beads) in units of mg of tracer per g of glass beads, divided by the aqueous concentration of tracer at equilibrium, in mg of tracer per mL of solution.

The laboratory batch method described in this section was taken from U.S. Environmental Protection Agency [1999a]. The objective was to determine the affinity of the RWT tracer for sorbing onto the surface of the glass beads. The experiment was performed using class 3 beads (see Table A-1). A single size class was used, since all glass beads used in this work were purchased from the same manufacturer, and were assumed to have uniform properties, apart from the differing grain diameters between classes. All

concentrations were derived from spectrophotometric measurements, using the linear calibration function (A.5).

A known mass of glass beads (M_{beads}) was added into each vessel (50 mL centrifuge tubes), and mixed with a solution of known volume V_{RWT} and concentration C_0 . The vessels were sealed and shaken continuously over a period of approximately 24 hours, to allow sufficient time for the dye to be adsorbed onto the glass beads. Following this equilibration period, the vessels were centrifuged for 10 minutes at a speed of 2800 revolutions per minute (rpm) to remove the fine particulate matter in suspension above the beads. The concentration C_i of the supernatant liquid was measured. The partition coefficient K_d was then calculated from [U.S. Environmental Protection Agency, 1999a]:

$$K_d = \frac{V_{RWT}(C_0 - C_i)}{M_{beads} C_i} \quad (A.6)$$

Its units are expressed in volume of solution per unit mass of porous medium (e.g. mL g⁻¹).

Triplicate K_d measurements were made using each of three RWT concentrations ($C_0 = 2.06, 4.12$ and 8.24 mg L⁻¹, respectively). For each value of C_0 , a vessel containing only the solution but no beads (container blank) was also used to assess whether any of the solution had adsorbed onto the container walls. For all samples, V_{RWT} and M_{beads} were equal to approximately 20 mL and 40 g, respectively. Results from the batch experiment are summarized in Table A-5.

Table A-5. Batch test results.

C_0 [mg L ⁻¹]	C_i [mg L ⁻¹]	V_{RWT} [mL]	M_{beads} [g]	K_d [mL g ⁻¹]
2.06	2.07	19.8104	40.6382	-0.001
	2.07	19.8869	40.7443	-0.003
	2.07	19.7907	40.7856	-0.003
4.12	4.16	19.9509	40.5155	-0.005
	4.24	19.9578	40.6155	-0.014
	4.24	19.8417	41.0509	-0.014
8.24	8.39	19.8119	40.7865	-0.009
	8.54	19.7710	40.4804	-0.017
	8.42	19.8670	41.0497	-0.010

Table A-5 shows that for all measurements there was no significant difference between the initial concentration and the equilibrium concentration; in fact, the equilibrium

concentrations were always slightly (but not significantly) higher. This indicates that RWT is not sorbed at all onto the glass beads.

It is worth noting that RWT has a high affinity for organic materials, such as microbial organisms. Several of the preliminary experiments conducted in the flow cell that were performed after a long period of stagnation (several weeks) showed significant retardation. The results were discarded based on the belief that the porous medium had suffered microbial contamination.

Optical properties

Rhodamine WT is a bright purplish-pink tracer, with a maximum absorption wavelength of 556 nm. Using a spectrophotometer, spectral absorption scans were performed for various concentrations. The results are shown in Figure A-11 for dye concentrations of 1.7, 4.2, 8.2 and 24.7 mg L⁻¹, which represent 0.5, 2.5, 5.0 and 15% of the stock solution ($C_{stock} = 164.7$ mg L⁻¹). Figure A-11 also shows that the absorbance spectrum of the camera filter (Kodak-Wratten filter no. 99, plotted as a thin dashed line) allows a maximum amount of light to be transmitted at wavelengths that correspond to RWT's maximum absorption wavelength (556 nm), shown as a vertical dashed line.

Spectral absorption for Rhodamine WT

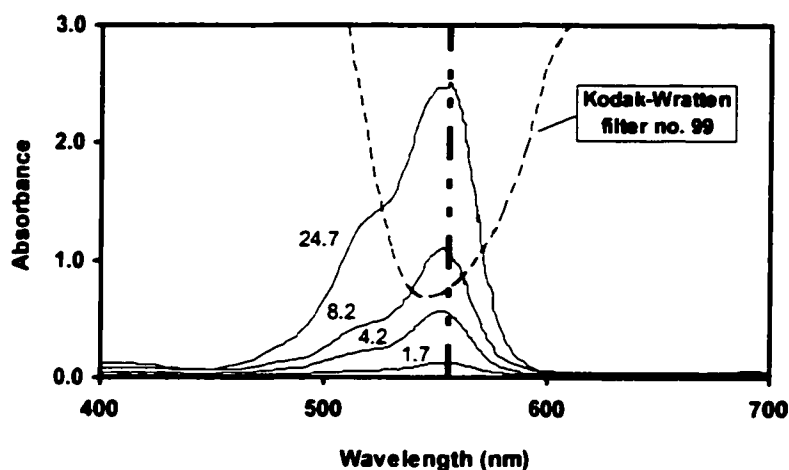


Figure A-11. Spectral absorption showing spectrophotometric characteristics of Rhodamine WT tracer and the Kodak bandpass optical filter used in the image acquisition system. The vertical line illustrates the maximum absorption wavelength of the tracer (556 nm).

4 IMAGE ACQUISITION SYSTEM

In this study, the flow cell was illuminated from behind using a custom-built light box, while images were captured from the front of the cell; the light was therefore transmitted through the flow cell. This “light transmission technique” was adapted from a study by *Tidwell and Glass* [1994]. It allows tracer plumes to be observed, integrated over their entire thickness. This method is in contrast with the more usual reflected light method where both the light source and the camera are on the same side of the flow tank. For instance, *Schincariol and Schwartz* [1990] and *Schincariol et al.* [1993] used a flow cell similar to the one used here, but the photographs they used in their work were taken using a standard front illumination. Because the glass beads they used – which we also use here – are not quite transparent, they were only able to observe the plumes along the front cell wall, down to a depth of a few bead radii at best. In general, transmitted light experiments offer a more accurate depiction of the tracer behaviour because the images obtained are integrated over the full thickness of the flow domain. On the other hand, reflected light experiments can be prone to substantial error caused by wall effects (accelerated flow at the walls due to the increased porosity in

their vicinity), or to local imperfections in packing. The source of error can be present in transmitted light experiments as well, but integration over the thickness of the flow cell tends to reduce their impact. This section presents the details of the image acquisition system used in this study, including the light source, the digital camera, and the choice of optical filter.

4.1 Lighting

One of the most important considerations during image acquisition is the light source. Image processing may help enhance or mask various features of an image, but cannot yield more information than is originally present on the image. Good illumination is essential to a good image, and care must be taken in selecting an appropriate light source. The light source used to illuminate the flow cell was designed with three key considerations in mind: brightness, uniformity and stability.

The *brightness* of the light source is extremely important in obtaining high quality images. Ample illumination is important in order to use smaller apertures because optical aberrations are enhanced by large lens apertures [Awcock and Thomas, 1995]. The brightness was maximized by aligning, side by side, ten high-luminosity fluorescent tubes inside a light box approximately 120 cm wide and 60 cm high. The light box was constructed from a standard industrial ceiling light fixture. The fluorescent tubes 50-watt Philips (colour temperature of 5500K). This particular type of fluorescent tube was selected because of its relatively narrow spectral emission (wavelength range), its even light distribution and low temperature radiation [Awcock and Thomas, 1995]. The light box was placed behind the flow cell, and opaque paper was used to cover those portions of the box extending beyond the cell to reduce glare. During the course of the flow and transport experiments, all lights in the laboratory were turned off, and the only illumination reaching the camera was from the back-illuminated flow cell. The *uniformity* of the incident light was maximized by lining the interior surface of the light box with crumpled aluminium foil, providing a large number of randomly oriented reflective surfaces to bounce light in all directions. Finally, a white translucent diffuser was placed between the light box and the flow cell. The resulting light source was both bright and uniform.

Because the basis of this study is to analyze and compare successive images, it is vital that the lighting conditions remain stable, that is, as constant as possible throughout the experiments. Variations in line current can lead to fluctuations in lighting intensity, degrading the *stability* of the illumination, which in turn can result in images of lesser quality. For this reason, the light source and image acquisition system were both powered by an uninterruptible power supply (UPS). This addressed some of the common problems associated with electrical power, including voltage surges, sags and line noise. Power surges are sudden increases in line voltage, while voltage sags (brownouts) are caused by periods of low voltage in utility lines. Line noise results from random fluctuations carried along with standard AC current.

4.2 Camera

The experiments described in this work were conducted using a monochrome Electrim⁴ digital camera. The camera is equipped with a charge coupled device (CCD) with a resolution of 1134 horizontal by 486 vertical picture elements (pixels). The sensing area of this solid-state image sensor, manufactured by Texas Instruments⁵ (model TC217), measures 8.67 mm by 6.59 mm. Thus, pixel dimensions are 7.8 μm by 13.6 μm . All images described in this work were acquired in interlaced mode, which effectively doubles vertical resolution by alternately scanning even and odd lines to produce a full image every second frame.

An 8-mm focal length C-mount lens, with manual aperture, provided the optics. The camera was connected to a personal computer, where all images were directly acquired and stored on the hard disk drive, via an interface card installed on the system bus. The image acquisition software allowed several parameters, including acquisition (exposure) time, bias and gain, to be adjusted until the best quality pictures were produced.

Much effort was devoted to testing the various combinations of settings for the light source, camera, and acquisition software prior to conducting each new set of experiments.

⁴ Electrim Corporation, 356 Wall St., Princeton, NJ 08540, (800) 683-5546

⁵ Texas Instruments Incorporated, 12500 TI Boulevard, Dallas, TX 75243-4136, (800) 336-5236

For each new packing of the flow cell, the optimal configuration was identified and these settings remained unchanged throughout the solute transport runs.

4.3 Camera filter selection

Careful adjustment of the various camera settings (e.g. gain, exposure time, bias and lens aperture) can dramatically enhance the contrast conveyed in pictures taken of the solute clouds; however, the contrast can be most significantly improved by eliminating the wavelengths from the transmitted light, except those maximally absorbed by the tracer. A pair of filters attached to the camera lens was used to “isolate” the tracer’s optimal absorption wavelength from the other wavelengths.

CCD sensors are quite sensitive to wavelengths in the near infrared, the segment of light just beyond the visible portion of the optical spectrum (700 to 1000 nm range). If no effort is made to prevent infrared light from reaching the CCD sensor, blurring or fogging of the image may occur [Russ, 1995]. To cut off infrared, a short wave band pass filter (Edmund Scientific filter P53,708) was mounted onto the camera lens to effectively absorb infrared light at wavelengths above 700 nm. The transmission spectrum of this filter is shown in Figure A-12.

Without any filtering the CCD sensors record the entire light spectrum over the 256 available shades of grey. Since the “white” light transmitted through the flow cell has a broad spectral output reaching beyond the visible portion of the optical spectrum, only a small portion of the available 256 shades of grey typically makes up the plume, thus resulting in poor resolution of the concentration distribution present in the image. In order to improve contrast, it is desirable to eliminate all the wavelengths from the incident light, except those maximally absorbed by the tracer.

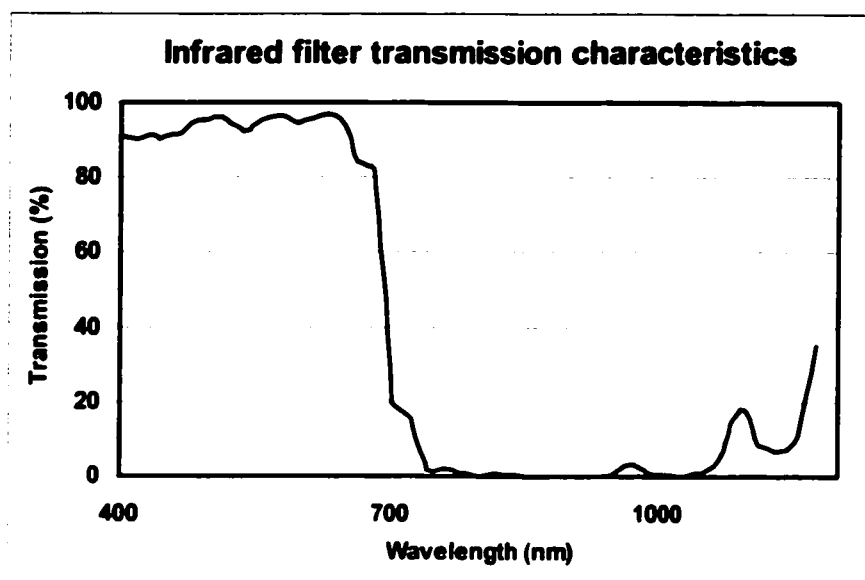


Figure A-12. Spectral transmission characteristics of infrared cutoff filter. The effect of this filter is to eliminate the effects of the presence of near infrared light incident upon the camera's CCD detector [from specification data provided by the manufacturer].

A band pass filter was also added to the camera's lens to enhance the contrast between the tracer and surrounding zero-concentration background (where only glass beads and water were present). Kodak Wratten filter no. 99 [Eastman Kodak Company, 1990], a gelatine interference filter with a peak transmission of approximately 545 nm (see Figure A-11), was used to isolate the portion of the visible spectrum near the tracer's peak absorption wavelength. This green filter was selected to match the tracer's maximum absorption wavelength (556 nm) by transmitting light only between approximately 500 and 600 nm.

The effectiveness of the light filtering system is shown in Figure A-13, by comparing an unfiltered image (Figure A-13(a)) to an image taken through the band pass filter (Figure A-13(b)). Concentrations below a minimum threshold cannot be distinguished from the zero-concentration background because of the slightly greenish shade of the glass beads, which also shows up in a light shade of grey.

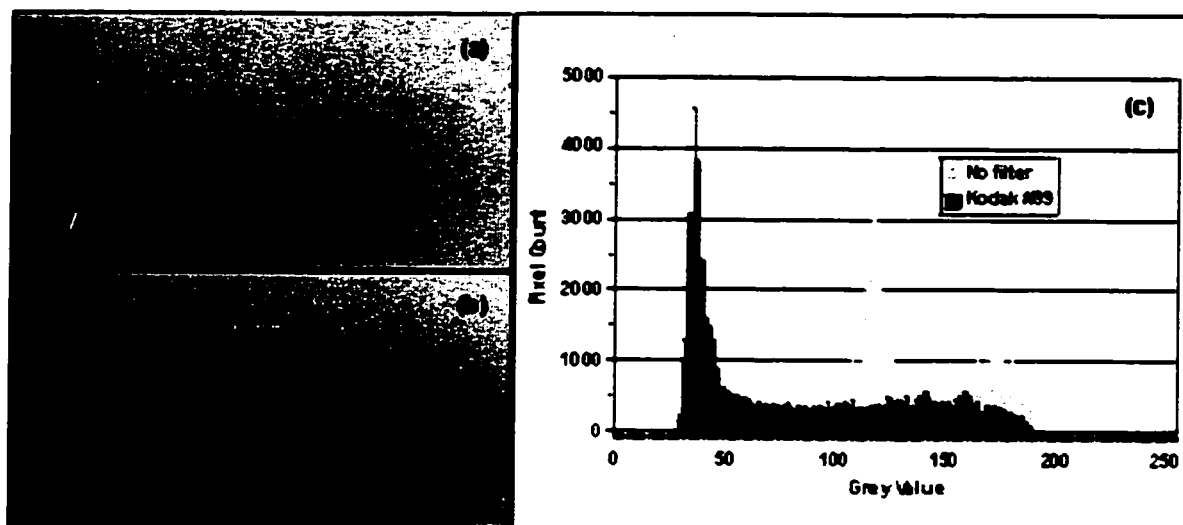


Figure A-13. Effect of band pass filter (Kodak Wratten #99) on image contrast. Histograms (c) showing the improvement in contrast achieved by using the band pass filter (Kodak Wratten No. 99), for a window 400 by 206 pixels². The range between minimum and maximum grey values (contrast) is enhanced by a factor of 2.7, from a range of 100 grey levels (99-199) for the unfiltered image (a) to an enhanced contrast with a range of 268 (27-295) with the filter (b).

APPENDIX B – Image Processing

“If I cannot picture it, I cannot understand it.”

– Albert Einstein

Images acquired with a high-resolution monochrome digital camera were used to obtain concentration profiles showing the time-space evolution through porous media of dissolved solute plumes. In order to extract information about the spatial and temporal distribution of the dissolved tracer, the images were processed to remove unwanted defects such as random noise, dark current, uneven illumination, or geometric distortions. Once the images were “cleaned up”, the information they contained – uncalibrated grey levels – was converted into units of solute concentration. For every scenario presented in the experimental portion of this work, the image processing procedure described here was used to produce a sequence of concentration maps ready for analysis using the technique of spatial moments described in Appendix C.

1 BACKGROUND

Although great care is used to control the conditions affecting image quality, corrections must be made to reduce unwanted variations introduced in the images by factors such as non uniform illumination, uneven bead packing in the case of the homogeneous porous medium, camera readout noise, or dark current. Principal sources of error are summarized in Table B-1, along with the methods used to correct them.

Table B-1. Factors and causes affecting image quality, and correction methods

Source of error	Principal cause(s)	Correction method(s)	Section
Random noise	Electronic circuitry	Signal averaging, Median filtering	3.1 3.3
Dark current	Thermal energy	Dark field subtraction	3.2
Spatial distortions	Non-square pixels, Camera misalignment	Image scaling, Image rotation	3.4
Uneven illumination	Light source, bead packing, imperfect CCD sensors	Normalization	4
Light source fluctuation	Power source variations	Standardization	5

The sequence of operations discussed in this section was applied to every image acquired during the course of the laboratory experiments. Each one of the image processing steps is summarized in Figure B-1 and discussed in detail in the following sections. The image processing procedure was subdivided into five sections, each with a specific function:

1. Image acquisition, to generate images of the highest possible quality
2. Corrections, to remove unwanted defects, including random noise, dark current and geometric distortions
3. Normalization, to minimize spatial variations in brightness
4. Standardization, to convert grey values into units of optical density and compensate for temporal illumination variations
5. Calibration, to convert optical density units into tracer concentrations

All image processing operations were performed using the public domain, open source program ImageJ⁶. Written in the Java programming language, two of the principal features of this software are that (1) it is independent of the operating system used (the most common being Microsoft Windows, MacOS and UNIX), and (2) it provides extensibility via Java plugins. These plugins provide unparalleled flexibility allowing non-standard processing and analysis operations to be performed on images.

⁶ National Institute of Mental Health, Research Services Branch, Bethesda, MD, <http://rsb.info.nih.gov/ij>

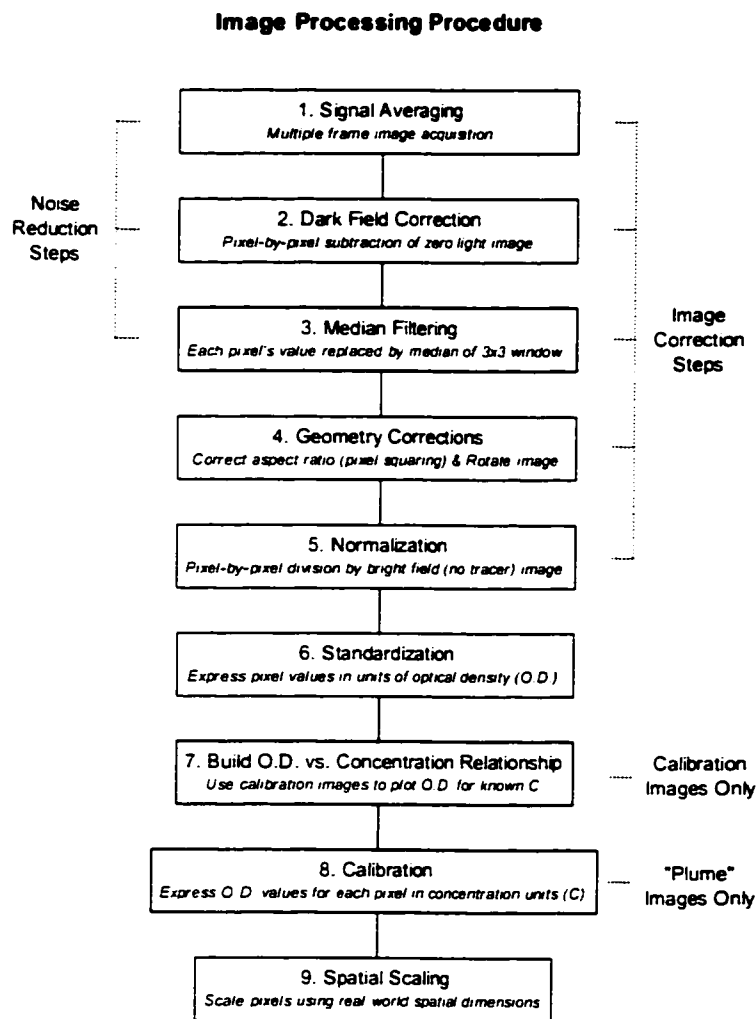


Figure B-1. Overview of the image processing steps. Steps 1 through 7 were applied to all images, while the procedures in steps 8 and 9 were necessary only for calibration or plume images, respectively. See text for details.

2 IMAGE ACQUISITION

One of the most critical aspects in producing images suitable for the analysis of a migrating tracer plume in the flow cell is image acquisition. As observed by *Russ* [1995], the use of image processing operations to improve the quality of images that have been stored on the computer depends on the quality of the original image. The acquisition procedure was therefore optimized for the set of conditions that prevailed in the laboratory.

The main factors of concern during this critical stage are (1) light uniformity and (2) readout noise. Although image defects introduced during acquisition cannot be completely eliminated, careful selection of the light source, combined with appropriate camera filters and optimized camera settings, are necessary to produce images of the highest quality. Image processing operations can greatly improve images, but care should be taken to make sure these are of the greatest quality possible in the first place.

Several aspects were addressed in order to maximize the quality of the images during acquisition, as described below.

2.1 Illumination

The light source used to illuminate the flow cell was designed with three key considerations in mind: brightness, uniformity and stability.

First, the *brightness* of the light box was made as high as possible by aligning, side by side, ten high luminosity fluorescent tubes inside an enclosure approximately 120 cm wide and 60 cm high. Next, *uniformity* of the incident light was maximized by lining the interior surface of the light box with crumpled aluminium foil, providing a large number of randomly oriented reflective surfaces to bounce light in all directions. The resulting light source was both bright and uniform.

The light box was placed behind the flow cell, and opaque paper was used to cover these portions of the box extending beyond the tank (to the left and right). During the course of the flow and transport experiments, all lights in the laboratory were turned off, and the only illumination reaching the camera originated from the back-illuminated flow cell. Black felt material was placed in front of the camera to eliminate reflections on the tank's front panel, and all images were acquired through an opening large enough to fit the camera lens.

Variations in electric line current can lead to fluctuations in lighting intensity, degrading the *stability* of the illumination, which in turn can result in images of lesser quality. The light source and image acquisition system were both powered by an

uninterruptible power supply (UPS), to address some of the common problems associated with electrical power, including:

1. Surges: sudden increases in line voltage, which are usually short in duration
2. Line noise: random fluctuations carried along with standard AC current
3. Brownouts (voltage sags): periods of low voltage in utility lines during periods of high consumption demand

2.2 Optical filters

While brightness, uniformity and stability of the incident light source are all factors affecting image quality, the use of optical filters, carefully selected to a particular set of conditions, can also dramatically improve image contrast. By isolating a narrow range of wavelengths, features whose colour falls within this window are enhanced, while objects outside this range will stand out less.

For the experiments described here, all images were acquired using two filters attached to the camera lens. The first filter, a band pass interference filter transmitting light only in the 500-600 nm range, was mounted in front of the lens. This Kodak Wratten No. 99 scientific grade filter [*Eastman Kodak Company*, 1990] was selected because its peak transmittance wavelength (approximately 545 nm) approached the maximum absorption wavelength of the Rhodamine WT⁷ tracer (556 nm). As shown in Figure B-2, this filter enhances the contrast significantly.

⁷ Crompton and Knowles Colors Incorporated, P.O. Box 33188, Charlotte, NC 28233, (704) 372-5890

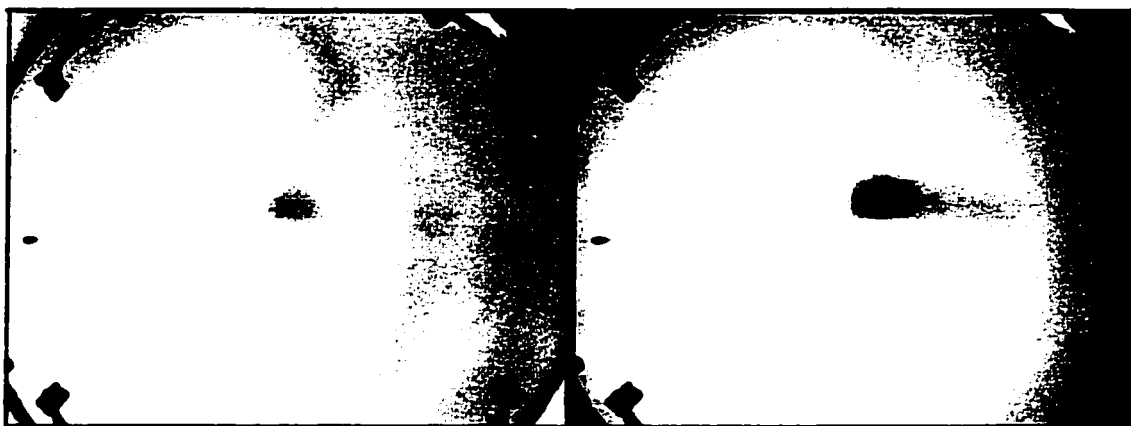


Figure B-2. Effect of a band pass filter on contrast of the solute plume. By matching the filter's peak transmission wavelength (545 nm) to the tracer's optimal absorption wavelength (556 nm), contrast is significantly enhanced. In the image acquired without a filter (a), the plume is quite bleak and the range of concentrations that can be observed is limited. On the other hand, the Kodak Wratten No. 99 optical filter (b) dramatically improves the resolution of the tracer.

The purpose of the second filter was to eliminate the blurring caused by infrared (IR) light. This IR-absorbing filter, according to the specifications supplied by the manufacturer (Edmunds Industrial Optics⁸), effectively absorbs wavelengths beyond the visible spectrum. This filter was used as a shortpass filter, allowing approximately 85-90% of the visible light (wavelengths in the 300-700 nm range) to be transmitted, while completely blocking out wavelengths longer than 700 nm.

Appendix A discusses the image acquisition system as well as the tracer (including plots of transmittance vs. wavelength for the filters and for the tracer).

2.3 Camera adjustments

Prior to each experiment, the camera's manual and software-driven controls were adjusted until an optimal combination of conditions was reached, to produce images of the highest quality and contrast.

⁸ Edmund Industrial Optics, 101 E Gloucester Pike, Barrington, NJ 08007-9897, (800) 363-1992

Lens aperture and focal distance

The 8-mm Cosmical lens used in our setup was selected because its field of view was large enough to accommodate the size of the flow cell, given the space available in the laboratory.

When adjusting lens aperture, a compromise must generally be struck between depth of field and brightness. Larger apertures (low focal ratio values – or f/number) allow a larger proportion of the incident light to reach the camera's detector, and are appropriate for dim settings, at the cost of a reduced depth of field. In this case, depth of field was irrelevant because of the two-dimensional nature of the experimental setup; a large aperture ($f/2.8$) was therefore used to maximize the amount of light passing through the lens.

In order to make images as sharp as possible, focal distance (focus) was adjusted until a minimum value of contrast was reached.

Anti-blooming, gain and bias

The camera software was used to enhance the sensitivity of the detector, while minimizing readout noise.

Blooming can occur when the individual sites on the charge coupled device (CCD) are saturated with more photons than they can collect. Charges can then overflow to affect the readout value in neighbouring pixels, resulting in artificial bright spots. The camera's anti-blooming feature was turned on at all times during image acquisition to minimize this phenomenon.

Camera gain can be adjusted to increase sensitivity of the detector. Gain was set as low as possible to minimize random noise induced by the electronics, yet high enough to produce images of high quality. Gain values between 190 and 200 were used throughout these experiments, but remained unchanged for any particular set.

The bias potentiometer was used to set the relative brightness level of the image, offsetting the effects of dark current, which is discussed in § 3.2. Bias was adjusted until

the minimum grey value in each image was as close to zero as possible. A bias of 100 was used for all experiments.

Exposure time

Finally, the last adjustment required for image acquisition was exposure time. Because solute plumes were not static, relatively short exposure times were required to minimize blurring caused by the motion occurring during the acquisition time for each frame. On the other hand, longer exposure times are desirable as they tend to reduce random noise, since over time this source of random error cancels out while the real signal continues to accumulate on the CCD (more on this shortly).

Exposure time was set at the beginning of each experiment, and remained unchanged throughout the duration of that particular run. With bias and gain values established as described above, the acquisition time for each image was determined prior to tracer injection so that maximum pixel values were close to – but without reaching – saturation. Exposure times, once determined, were used throughout the full duration of a particular experiment. Values used were between 230 and 255 ms (homogeneous medium) and between 100 and 120 ms (heterogeneous medium).

3 CORRECTION

The image correction procedure can be subdivided into four main components: (1) Signal averaging, (2) dark field correction, (3) median filtering, and (4) geometry corrections. Each is explained in detail below.

3.1 Signal averaging

The image acquisition chain is made up of a number of electronic circuits and wires that contribute to degrade the signal by introducing random noise. Depending on the conditions specific to a particular experiment, each image was acquired over a period of $1/10^{\text{th}}$ to $1/4^{\text{th}}$ of a second (100 to 255 ms). This integration period is long enough to

remove a portion of the random noise, based on the principle that unwanted noise gradually cancels out while the actual signal accumulates.

Additionally, random noise can be reduced by averaging static images over multiple frames, therefore improving the quality of the image. In theory, the signal-to-noise ratio increases at a rate equal to the square root of the number of frames [Russ, 1995].

In this work, all static images (all images except those where a migrating tracer plume was present) were averaged over four frames, resulting in a two-fold improvement in image quality. This was applied to all dark field (zero light), bright field (no tracer) and calibration (tracer-saturated) images.

Figure B-3 illustrates how frame averaging improves image quality by comparing the histograms of grey values over a relatively uniform portion of the porous medium. Pixel values were obtained from a 200 x 200 window near the centre of a bright (no tracer present) field image (run C3.ss12.f). The minimum, maximum, mean and mode are virtually unchanged by the frame averaging process, while standard deviation is lowered by 3.4%, from 5.05 to 4.88. Close examination of the two histograms clearly shows a reduction in the amplitude of local ups and downs in the averaged image (Figure B-3(b)), compared to the single-frame image (Figure B-3(a)).

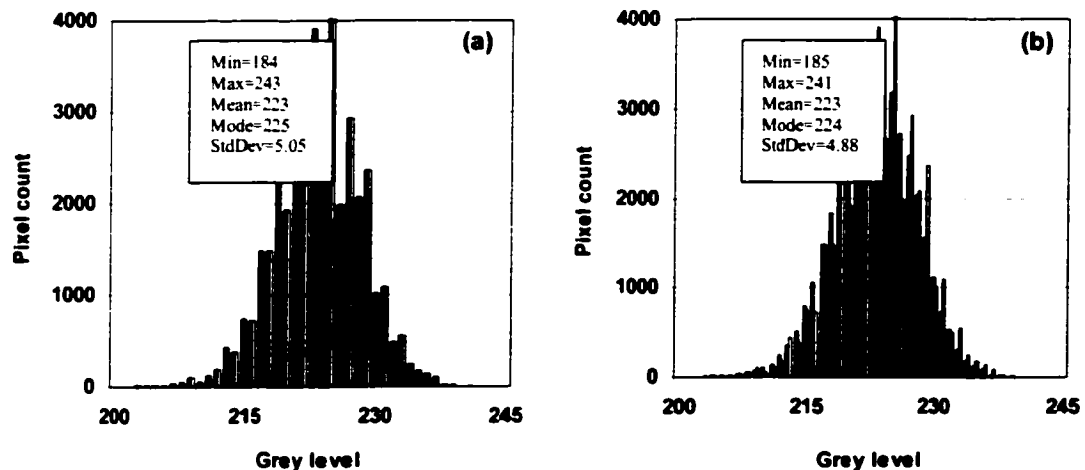


Figure B-3. Effect of frame averaging on random noise, shown by comparing histograms for a single frame (a) and a composite image averaged over four frames (b).

3.2 Dark field correction

When light is completely blocked out from the camera's CCD sensor, a brightness value of zero (pure black) should be recorded at each pixel. However, the CCD's internal circuitry generates thermal energy that induces a voltage (dark current), which is in turn digitized into a brightness value. Dark current refers to the artificial brightness of the acquired image that does not correspond to light reaching the detector. This phenomenon increases with increasing temperature of the CCD, and the visible effect of the dark current will increase with longer exposure time [*Electrim Corporation*, 1994].

The most efficient means of reducing dark current is by cooling the CCD chip in order to reduce thermal noise. This can be accomplished by attaching a Peltier cooler to the sensor [*Russ*, 1995]. However, such a device was not available to conduct the experiments described here, and the presence of a certain amount of thermally induced dark current was unavoidable.

Contribution of the dark current can be removed from all images by subtracting, pixel by pixel, a dark field image of comparable exposure time [e.g. *Baldock and Poole*, 1993]. This correction was performed on all images. For any given pixel, the dark current corrected grey level is given by:

$$\begin{aligned}(g_B)_i &= (g_B - g_D)^{RAW} \\ (g_C)_i &= (g_C - g_D)^{RAW} \\ (g_P)_i &= (g_P - g_D)^{RAW}\end{aligned}\tag{B.1}$$

where g refers to the grey level, i is the pixel's index, and subscripts B , C , P and D are used to distinguish between bright field, calibration, plume or dark field images, respectively. Note that the designation *RAW* is used indiscriminately for images averaged over multiple frames as well as for single-frame images.

Figure B-4 shows a typical dark field image that was averaged over four frames, and acquired while the camera's aperture was closed to prevent any light from reaching the CCD. Note that for illustrative purposes, the contrast from the original image was increased to show the variations in dark current across the image. For illustrative

purposes, the range of pixel intensity values in the actual dark image ($g_{min} = 5$ to $g_{max} = 26$) was thus expanded to occupy the full range of 256 available grey values, from $g_{min} = 0$ (black) to $g_{max} = 255$ (white). Figure B-4 shows a uniformly distributed array of artificial pixel values caused by dark current.

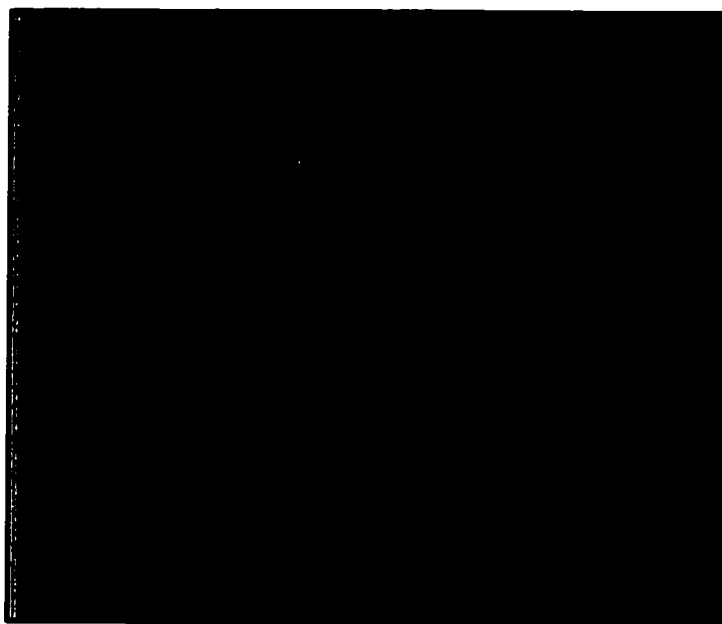


Figure B-4. Typical dark field image with contrast expanded to show the thermally induced dark current, which translates into an artificial signal. Despite the fact that this image was acquired with the lens aperture closed, all pixels have a non-zero value, with intensities between $g = 5$ and 26), resulting from the dark current.

3.3 Median filtering

As described in § 3.1, signal averaging reduces, at each pixel, the noise randomly distributed in each image (known as white noise). Although it is reduced, the random noise is not completely eliminated in the averaged image. Signal averaging was used in static images but not in images of migrating tracer plumes because of the need to reduce artificial smearing of the concentration distribution that results from the unavoidable delay between acquisition of the individual frames. The migrating tracer images therefore contained a greater amount of white noise than static images.

The remaining white noise may be reduced further by applying a median smoothing filter. This filter will also reduce the effects of defects such as very small (opaque) impurities present in the porous medium or small scratches on the walls of the flow cell. This operation consists in replacing the intensity (grey level) in each pixel with the median of intensities from a neighbourhood surrounding and including the pixel. This local window is moved across the entire image and the median value obtained at each location replaces the grey level from the original image. Median smoothing is more appropriate than averaging, because it is not susceptible to extreme values and will preserve the location of boundaries or concentration distributions [Russ, 1995]. This is illustrated in Figure B-5.

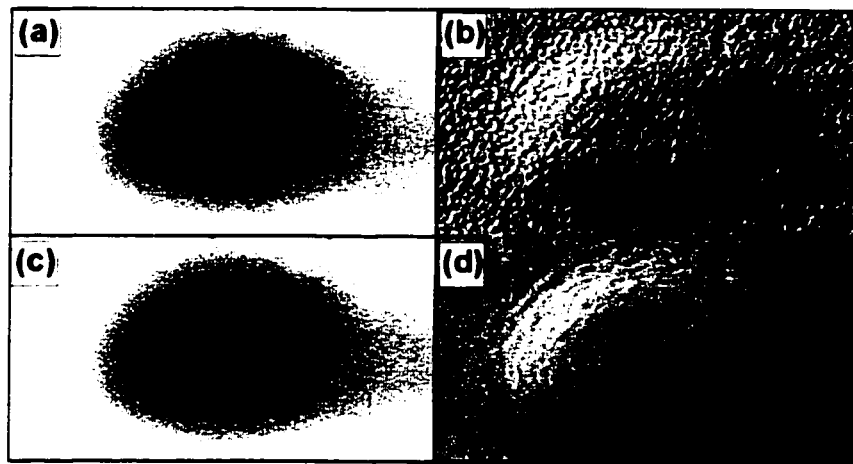


Figure B-5. Effect of median filtering on small-scale noise level. The original solute plume (a) is shown with its relief map (b) to help visualize random noise. Applying a 3x3 median filter dramatically improves image quality, to produce a smoother plume (c); this is seen more easily in the relief map (d).

Although neighbourhood patterns of various sizes and complexity are possible for median filtering, we used a local window size of nine pixels (3 by 3) for all images. The local area of influence was chosen to smooth out high-frequency noise and defects, while preserving the larger features (solute plumes).

3.4 Geometry correction

Individual sensors on our digital camera's CCD are 7.8 μm wide by 6.8 μm high. Since pixels displayed on a computer monitor are square, it is necessary to correct for the rectangular aspect ratio to preserve the geometry and accurately display images on screen. Without pixel squaring, images would be artificially stretched in the vertical direction leading to inaccurate spatial measurements. Calculations that use pixel values, such as spatial moments (see Appendix C), are simplified if pixel dimensions in the horizontal (x) and vertical (y) dimensions are equal.

Each image, following frame averaging (when applicable), dark field correction and median filtering, was transformed using a bicubic interpolation algorithm from its original dimensions of 1134 by 972 pixels², to a new image size of 1134 by 847 by reducing the height by a factor of (6.8/7.8). This operation was successful in correcting for the CCD's width:height aspect ratio.

4 NORMALIZATION

Overall lighting conditions can fluctuate in time, in response to variations in the building's power output as demand varies throughout the day; and also in space, mostly because of inhomogeneity in the light source and in the porous medium's packing density or grain size distribution (especially when the porous medium is heterogeneous).

Normalization corrects for temporal and spatial variations in brightness by expressing each pixel in images containing the tracer as a ratio relative to a bright field image, acquired prior to each experiment with only water and the porous medium present in the flow cell (no tracer). This effectively eliminates systematic, non-random brightness variations.

Pixel values (indexed on i) are normalized for images containing a solute plume ($g_p^\bullet$) _{i} and for calibration images ($g_c^\bullet$) _{i} using:

$$\left(\dot{g}_P \right)_i = \left(\frac{g_P}{g_B} \right)_i \quad (\text{B.2})$$

$$\left(\dot{g}_C \right)_i = \left(\frac{g_C}{g_B} \right)_i \quad (\text{B.3})$$

where, as before, g refers to the grey level, and the subscripts P , B and C refer to the bright field, calibration and plume images, respectively. Normalization produces an image that only shows the difference between an image containing the tracer, and the corresponding bright field image containing only the porous medium and water. This step is very useful for removing background brightness variations or shifts in incident light intensity fluctuations. In addition, normalization constitutes a critical step for heterogeneous media because it effectively removes brightness variations, caused by the non-uniform optical properties of porous media of varying grain sizes.

Figure B-6 illustrates the effects of the normalization procedure for images containing a migrating solute plume, taken from two separate experiments. In the first case (top row, Figure B-6 (a) to (c)), the plume image is from an experiment within a homogeneous porous medium, while images from the bottom row (Figure B-6 (d) to (f)) were generated during an experiment where the tracer was migrating through a heterogeneous medium.

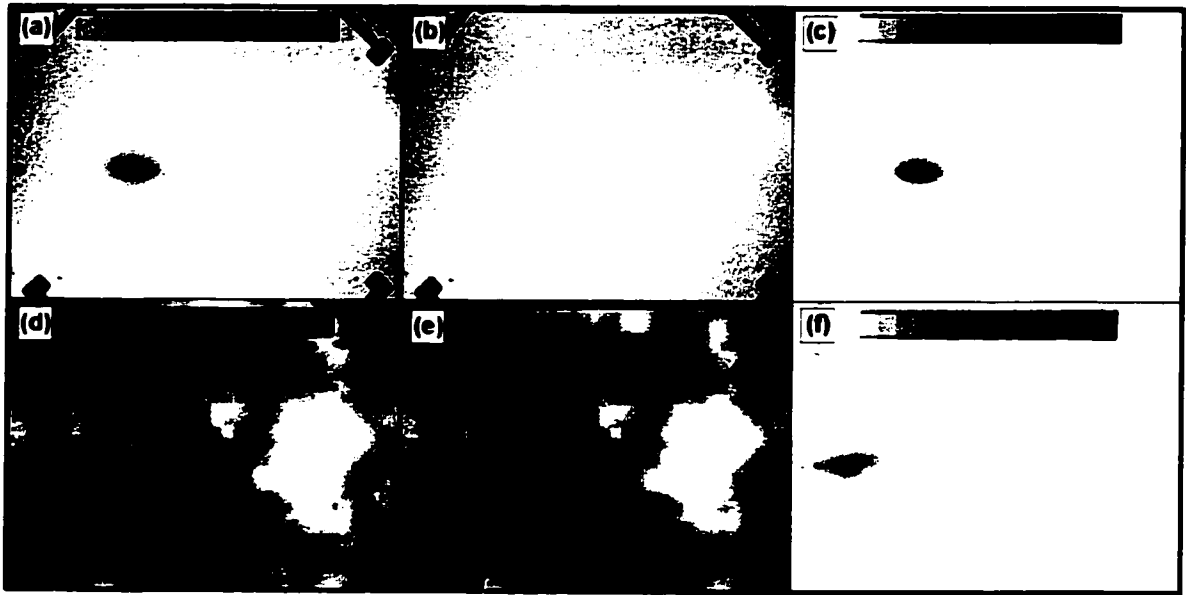


Figure B-6. Effect of normalization. An image containing a plume migrating through a homogeneous porous medium (a) is divided pixel by pixel by the bright field image (b). In the resulting normalized image (c), the zero-concentration (background) is more uniform. In the case of a heterogeneous porous medium (d), it is very difficult to distinguish the solute plume from the background. After division by the bright field image (e), the resulting normalized image (f) shows the tracer distribution very clearly. Each image is 52.8 cm wide by 39.5 cm high.

Because additional random noise is introduced by the normalization operation (division), normalized images were filtered a second time using a 3x3 median filter, as described previously (see Section 3.3).

5 STANDARDIZATION

Following correction (Section 3) and normalization (Section 4) to remove unwanted defects and features, images containing a migrating tracer cloud were converted so that their pixel values (grey levels) were expressed into units of optical density D . This operation – standardization – produces for each image a function that converts grey values into density units. This step is necessary because there is no easy way to know the exact relationship between optical density and pixel brightness, and this function must be determined empirically.

The process of relating pixel grey levels to a set of density standards is sometimes referred to as “calibration”. To avoid confusion, the term “standardization” is used instead, reserving the word “calibration” for the next topic of discussion where pixel values are converted into units of solute concentration (see Section 6).

5.1 Density standards

Images can be standardized to optical density units using step wedges. Step wedges are transparent tablets made of glass or acetate-like material, and subdivided into a number of linearly increasing calibrated optical densities. Linear stepped density filters are available commercially (e.g. from Edmunds Industrial Optics⁹), but their cost can be prohibitive (approximately CAD \$750). When calibration is performed using such a tablet, images can be expressed in terms of “true” optical density (O.D.) units, which is a measure of the proportion (I) of incident light (I_0) that passes through the sample without being absorbed or scattered [Russ, 1995]:

$$O.D. = \log_{10} \left(\frac{I}{I_0} \right) \quad (B.4)$$

For the experiments described here, a standard scale was constructed inexpensively using a sheet of light-coloured theatrical gelatine (plastic film) normally used to alter the colour of spot lights. The plastic film was cut into strips (2.54 cm wide by 3.81 cm tall) that were stacked and offset successively to produce a stepped tablet (or step wedge) wherein each new layer added a constant optical density to the previous step. The optical density increments varied from a single layer to 14 layers in the darkest optical density. We selected a light-pink theatrical gelatine film that provided good contrast with the camera filter (see Section 2.2 for a discussion on optical filters). The actual optical density of the translucent step tablet was not measured because it was used as a consistent standard scale that was present in all images. The step wedge can be seen at the top of each image (e.g. Figure B-6).

⁹ Edmund Industrial Optics, 101 E Gloucester Pike, Barrington, NJ 08007-9897, (800) 363-1992

Since the true OD of the material used to build the standard scale was not measured, pixel grey values in standardized images were expressed in terms of “gelatine optical density”, or simply “density” (D). An arbitrary value of $D(1) = 1.00$ was assigned to the first step (a single layer), and the densities of the other squares on the standard scale were therefore equal to $D(2) = 2.0$, $D(3) = 3.0$, and so forth, up to a maximum value of $D(14) = 14.0$ for the last square. The optical density of the darkest square therefore corresponds to the highest tracer concentration that can be used, and must therefore be at least as dark as the undiluted tracer injected into the porous medium.

For any given image, the normalized grey values g_i^* were averaged over all pixels within each of the 14 squares of the standard scale. The resulting table of measured (averaged) values $\overline{g^*}(l)$ vs. known $D(l)$ standards, where $l = 1, 14$, was used to compute the standardization function as described in the following section. A typical standardization table, generated from the image in Figure B-6 (c), is presented in Table B-2.

Table B-2. Standardization data (from the image in Figure B-6 (c)) showing recorded mean grey values ($\overline{g^*}(l)$) for each standard square l of known density $D(l)$. The standard deviation, mode, minimum and maximum are also shown for each tabular entry. Note that the zero-density measurement $D(0)$ was averaged over a region located directly below the standard scale.

l	$D(l)$	$\overline{g^*}(l)$	Standard deviation	Mode	Minimum	Maximum
0	0.00	250.23	1.44	250	243	255
1	1.00	209.49	1.90	210	198	215
2	2.00	174.10	1.55	175	169	179
3	3.00	145.72	1.53	146	141	150
4	4.00	121.36	1.61	122	117	128
5	5.00	103.40	1.44	104	99	109
6	6.00	87.00	1.01	87	84	91
7	7.00	74.28	1.46	74	70	79
8	8.00	63.46	1.19	63	60	68
9	9.00	54.33	1.48	54	49	58
10	10.00	46.44	1.09	46	43	50
11	11.00	39.67	1.15	40	37	43
12	12.00	35.53	1.30	36	31	39
13	13.00	32.17	1.28	32	27	36
14	14.00	29.03	2.44	28	18	41

5.2 Rodbard standardization function

For each image acquired in the course of an experiment, the grey value of each pixel needs to be expressed as an optical density in order to remove unwanted effects due to power fluctuations, etc. A standardization function must therefore be generated from the $\overline{g^*}(I)$ vs. $D(I)$ data for each image. This was achieved using the powerful image processing software ImageJ (IJ), available for download from the National Institute of Mental Health (NIH)¹⁰. IJ was also used to calculate spatial moments from images, using a plugin written specifically for this project (see Appendix C for a complete description of this plugin, including its source code). Although there are many curve fitting methods to choose from in IJ (e.g. linear, exponential, polynomial, or logarithmic), the Rodbard function was selected over other functions because it consistently produced curves with the highest degree of fit, with typical R^2 values above 0.99 and often as high as 0.999.

The Rodbard is a four parameter general curve fit function developed by David Rodbard at NIH. It is of the form:

$$y = c \cdot \left[\frac{(a - x)}{(x - d)} \right]^{1/b} \quad (\text{B.5})$$

where a , b , c and d are curve fitting parameters calculated from the tabulated standardization data ($\overline{g^*}(I)$ vs. $D(I)$). The independent variable x generally represents grey levels, while the dependent variable y is in the same units as the standard used ("Gelatine Optical Density" (D) units in this case).

To illustrate the application of the Rodbard Equation to standardize images, the procedure is outlined below for the image presented in Figure B-6 (c). A standardization function was fitted to the data from Table B-2 using the Rodbard curve fitting function. The resulting equation is:

$$G = 0.000827 \cdot \left[\frac{(19.011105 - x)}{(x - 517.858680)} \right]^{1/0.261253} \quad (\text{B.6})$$

The function and measured data from which it was generated are plotted in Figure B-7.

¹⁰ National Institute of Mental Health, Research Services Branch. Bethesda, MD, <http://rsb.info.nih.gov/ij>

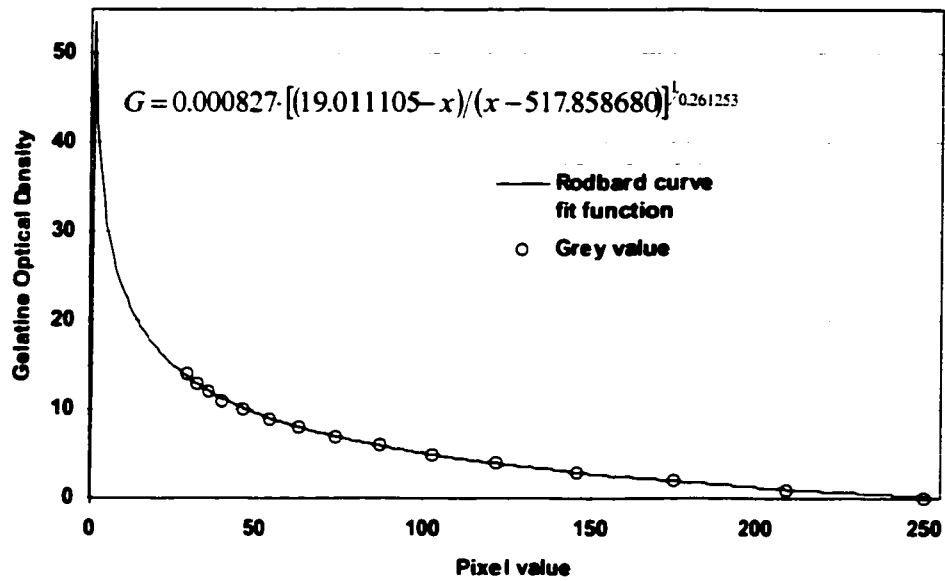


Figure B-7. Rodbard curve fit function used to standardize the image from Figure B-6 (c). Goodness-to-fit $R^2 = 0.998788$ and standard deviation $s = 0.123352$.

The effects of standardization are shown in Figure B-8, which is a plot of grey scale values before and after standardization as a function of position along the step wedge. The relationship between grey values prior to standardization and optical density of the gelatine (showing up as a series of plateaus) is non-linear (dashed line on Figure B-8). On the other hand, the relationship in the standardized image is linear, showing the increments of $D = 1.0$ for each square of the step wedge.

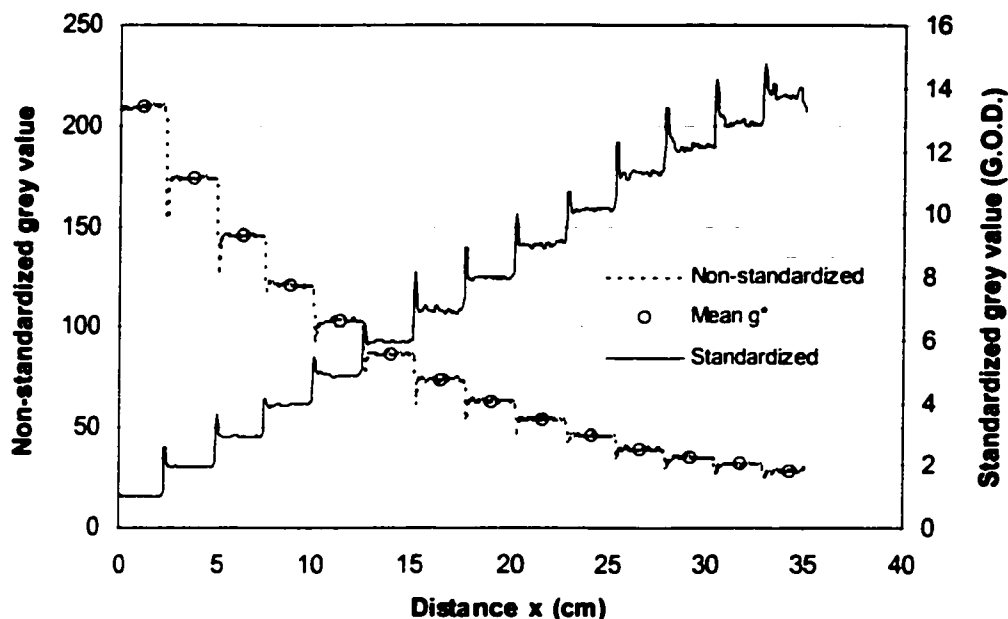


Figure B-8. Effect of standardization showing the pre-standardization grey values (dashed line) and standardized values (solid line). Circles represent the values averaged over each square on the step wedge, that were used to derive the standardization function in Figure B-7. Pixel values were averaged in the vertical direction over the entire height of the step wedge (approximately 3 cm, or 65 pixels). The effect of standardization is to linearize the relationship between grey scale and optical density. Note that random noise shows up as minor variations in grey value for each square on the scale. The more pronounced peaks are introduced by dark lines that were drawn to delineate each square, and are not taken into account when computing the standardization function.

Because concentration is directly proportional to optical density, solute concentrations present in standardized images can be analyzed to obtain quantitative measurements about the spatial distribution of the solute. Standardized images, where pixel values are expressed in units of optical density, can thus be used directly to analyze the solute's spatial distribution, where concentration of the solute will be expressed relative to the initial concentration.

6 CALIBRATION

Because it is not practical to include a set of internal standards of known concentrations in the flow cell without disturbing the flow field, calibration was performed in a two-step

process. First, images were standardized (previous section) using an external standard (stepped tablet) placed on the flow cell and present in all except bright field images. Next, standardized images were converted from units of optical density – or in this case, in “gelatine optical density” units – into units of solute concentration, a process coined as “calibration”.

6.1 Relationship between light absorption, optical density and concentration

Absorption can be defined as the amount of incident light that is prevented from reaching the camera detector by translucent objects located between the light source and acquisition apparatus. The radiation (energy emitted by the light source) is selectively absorbed according to the chemical composition of the objects in its path [Russ, 1995]. In this study, light absorption is due mainly to the porous medium and to the RWT dye. Normalization (described in §3) effectively removes the contribution of the porous medium to overall absorption, producing images containing only the solute and the step wedge. Following standardization to a set of internal standards (see §4), pixel brightness values are expressed in units of optical density, and can be used to interpret the tracer’s concentration distribution. For this purpose, it is necessary to use external standards (solutions of known tracer concentration) to determine the function that relates the optical density measured in the flow cell (G_C) to the known concentration (C_{cal}). This is the objective of the image calibration step, described in this section.

Dye concentration and absorption (a measure of optical density) are related linearly according to Beer’s law. This relationship is illustrated in Figure B-9, where the absorption at a wavelength $\lambda = 556$ nm was measured using a spectrophotometer for tracer standards, with concentrations ranging from zero (pure water) up to 16.47 mg L⁻¹.

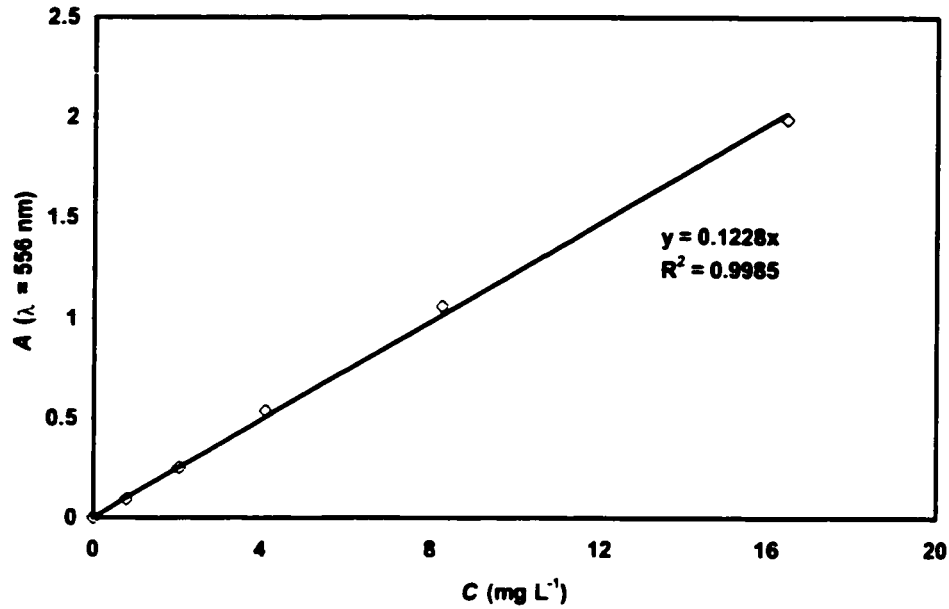


Figure B-9. Spectrophotometric absorption data for various tracer concentrations ($\lambda = 556$ nm), showing the linear relationship between A and C .

Because this relationship is linear, only a few calibration images are required for a given porous medium to derive the optical density vs. concentration (G_C vs. C_{cal}) relationship.

6.2 Concentration calibration (G_C vs. C_{cal})

Calibration was done by temporarily flooding the region of interest (ROI) in the flow cell with tracer solutions of known concentration and measuring the optical density G_C , averaged over the ROI. This was accomplished by supplying the reservoirs connected to ports nos. 9 and 10 (adjacent to the injection port) with the solution of desired concentration C_{cal} rather than water. Limiting the tracer flooding to these two ports prevented the tracer from invading the flow cell where the step wedge was located. The ROI is the central portion of the flow cell, approximately 50 cm wide by 30 cm high, where the migration of tracer plumes was observed during the flow experiments. It almost occupies the full width of the tank, but only extends approximately between the handles on the tank cover, which show up as opaque objects in the images (e.g. see

Figure B-6). A typical calibration image is shown in Figure B-10, for a homogeneous porous medium and $C_{cal} = 5.17 \text{ mg L}^{-1}$.

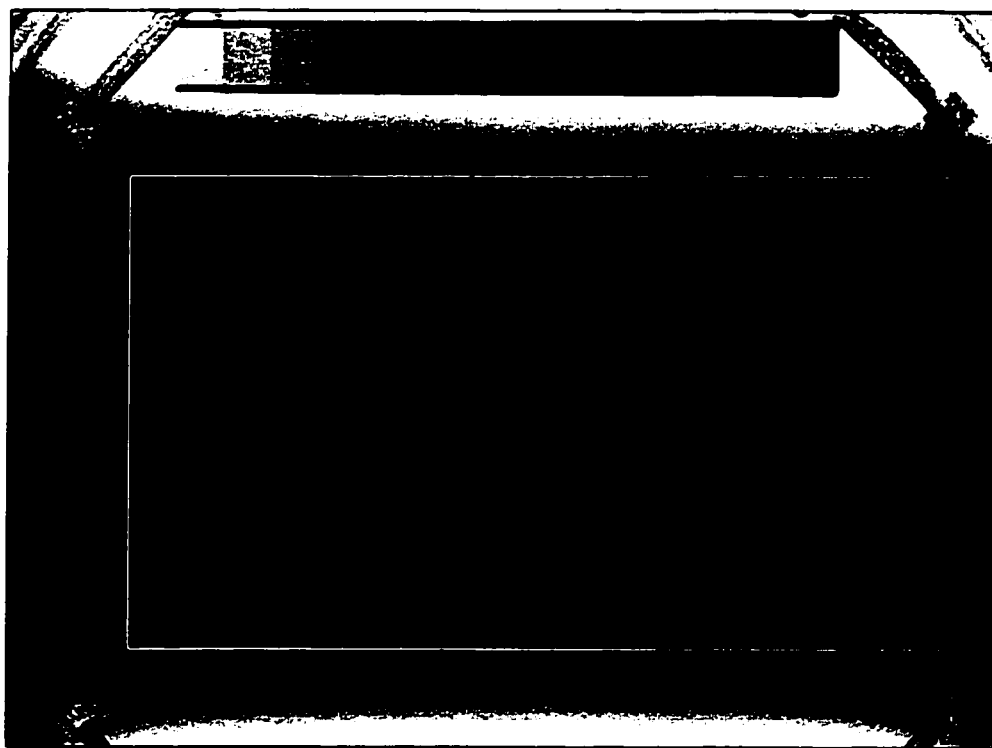


Figure B-10. Typical calibration image following correction, normalization and standardization steps. The tracer solution ($C_{cal} = 5.17 \text{ mg L}^{-1}$) was introduced in the flow cell through upgradient ports nos. 9 and 10, and allowed to saturate the region of interest (ROI). The mean value of $D = 13.03$ (standard deviation = 0.44) was measured over the ROI, shown in white.

Although only two calibration images are technically sufficient to derive the linear relationship G_C vs. C_{cal} , it is preferable to use several images, each with a different value of C_{cal} , in order to reduce the uncertainty associated with the calibration function.

One such calibration image (averaged over four frames) was obtained for each of four calibration concentrations C_{cal} . Each image was then corrected, normalized and standardized using the internal standard (step wedge), as described previously, and the pixels' grey values were expressed in units of optical density. Each calibration image thus produced a single point on the calibration curve, where the normalized grey level G_C ,

averaged over the ROI, was plotted as a function of the known concentration C_{cal} (see Figure B-11). For calibration purposes, four dye concentration values were used in the calibration: 5.17, 1.91, 0.82 and 0.00 mg L⁻¹ (from a background image containing only porous medium and water).

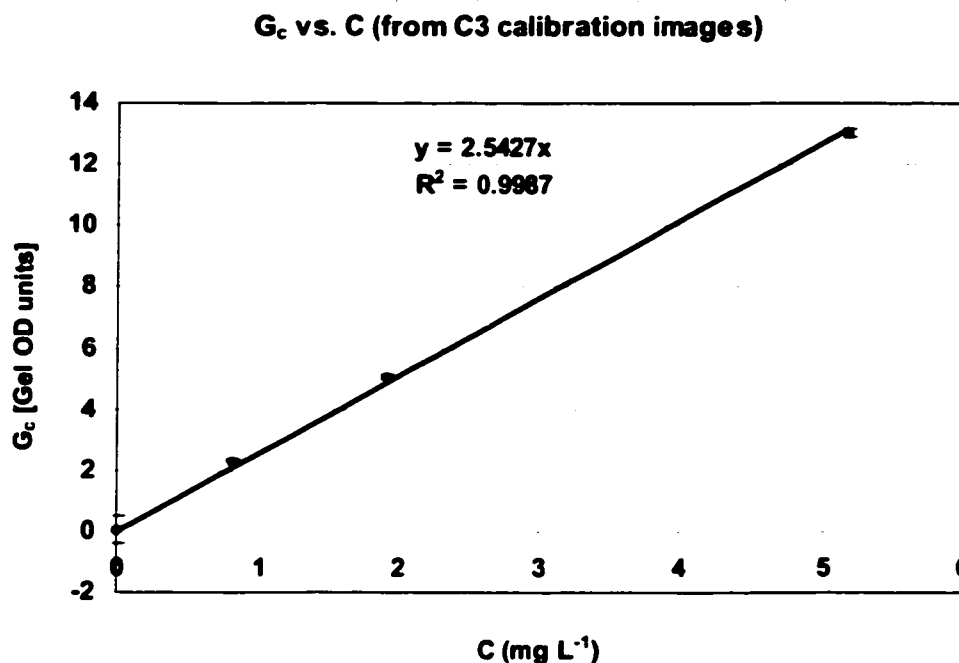


Figure B-11. Calibration curve for homogeneous porous medium. Each point on the graph was obtained from a single calibration image, by averaging the standardized grey values (in units of Gel OD) over all the pixels in the ROI. Tracer concentration of 0.82, 1.91, and 5.17 mg L⁻¹ were used. The zero-concentration point was measured from an image of the flow cell containing only the porous medium and water.

Applying the linear calibration function derived from Figure B-11 to image sequences containing a solute plume, however, proved to be problematic. A gradual increase over time of the total dissolved mass estimated from “calibrated” images suggested that early time concentrations were somewhat under-predicted, resulting in a total mass that was lower than predicted from the known injected mass. Although the cause for this is not clearly understood, it is likely an artefact of the acquisition and/or processing of images. For example, it might be attributed to the fact that the bright pixels surrounding the solute

plume (where no tracer is present) artificially brighten the plume, in effect “washing out” the plume area; because at early times plumes are smaller, this phenomenon presumably influenced the results by a greater amount shortly after tracer injection, to gradually become less apparent as the migrating plume spread out and occupied a larger portion of the flow cell. Late time (diluted) solute plumes are therefore believed to produce a better approximation of the actual mass in solution.

As an alternative, an empirical calibration procedure was developed in order to circumvent this problem and produce concentration distributions from image sequences, where total mass was constant over time. First, the concentration distribution was predicted from a numerical simulation reproducing the experimental conditions from the simplest scenario (Case 1, Run 1). Note that the numerical model’s longitudinal and transverse dispersivities ($\alpha_L = 0.015$ cm and $\alpha_T = 0.0015$ cm, respectively) were obtained by calibration to the actual plume from the image sequence. Second, the grey values from standardized images were corrected using a scaling factor equal to the ratio between the maximum values (peaks) from a “late time” ($t = 25$ min) plumes (laboratory vs. simulation). The “late” plumes were selected based on the assumption discussed above that the estimate of total mass from images gradually improves over time. A correction factor equal to 0.692 was used for all images ($\gamma = 0.692 G$). Third, a third-order polynomial function was fitted through a scatter plot of the corrected grey values γ (from images) against the predicted concentration values (from the numerical simulation). The “calibration” equation is given by:

$$C = a\gamma^3 + b\gamma^2 + c\gamma + d \quad (\text{B.7})$$

where the factors were adjusted empirically until the total mass was relatively constant over time. These were $a = 0.06$, $b = -0.2$, $c = 1$ and $d = 0$.

6.3 Spatial scaling

Pixel dimensions were specified for each image by using fiducial points marked on the flow cell. For this purpose, the number of pixels n separating the left and right edges of

the standard scale (of known length $l = 35.56$ cm) was determined for each experiment. Pixel size s was obtained from:

$$s = l/n \quad (B.8)$$

Spatial scaling was identical for all images because the same lens was used throughout, and also because the distance between flow cell and camera was held constant. With the length of the standard scale ($l = 35.56$ cm) and number of pixels ($n = 757$) both being constant, pixel dimensions were therefore equal to $s = 0.047$ cm (21.29 pixels per cm). Because image dimensions were modified to make pixels square (see §2), the scale was identical in both the horizontal and vertical directions.

Thus, the spatial resolution for the experiments presented in this work was on the order of less than half of a millimetre, resulting in a concentration distribution that can be considered continuous. This high resolution is at least one order of magnitude finer than what could be achieved using a standard setup that uses sampling ports, with the added benefit of providing an instantaneous snapshot of the tracer plume without disturbing the migration patterns nor stopping flow.

7 CONCLUSIONS

The procedure presented in this section provides a fast, inexpensive, non-intrusive, non-destructive and accurate method of measuring tracer concentrations from images of the solute's distribution in a flow cell saturated with flowing water. In the context of our laboratory experiments, this technique provides us with an efficient way of characterizing the spatial distribution of a migrating cloud of coloured tracer solution, and of following its evolution over time.

This procedure is applicable to heterogeneous porous media, where beads of various sizes are used, because spatial variations in shading due to variations in the optical properties of the porous medium are removed by the normalization process. This also accounts for uneven bead packing, illumination and/or efficiency of the CCD array.

APPENDIX C – Spatial Moments

In their pioneering study of the dispersion of a solute dissolved in a fluid flowing through a tube, *Aris* [1956] demonstrated that spatial moments of concentration distributions can provide a useful representation of the motion of solutes, integrated over a macroscopic region. Because of their suitability for describing the motion of a dissolved solute in field, laboratory or numerical solute transport investigations, spatial moments constitute a valuable quantitative tool for monitoring the dispersive behaviour of solutes in porous media.

In the present study, experimental evidence consisted of two- and three-dimensional spatial distributions of solute concentrations produced during laboratory and numerical experiments. Several presentation methods can be used to conveniently summarize and display the solute distributions for analysis. These tools may take the form, for example, of charts, contoured concentration maps or, in the case of laboratory experiments, photographs of the solute clouds. Although such visual representations of the data are usually quite successful to provide an intuitive picture of the tracer cloud distribution and evolution, they cannot easily be used to compare plume characteristics across experimental scenarios; and they are, at best, qualitative.

The following sections provide the theoretical background used to derive the spatial moments from spatial distributions. Although the equations presented here are applicable to sets of numbers from a variety of contexts, the emphasis is placed on their use in describing dissolved solute plumes migrating through porous media.

1 BACKGROUND

Spatial moments (also called “statistical moments”, or “moments of inertia”), constitute a quick and convenient method of quantitatively summarizing a dataset whose central

tendency is sufficiently important [*Press et al.*, 1992]. The most commonly used examples of spatial moments are the sum (zero-order), the mean (first-order) and the variance (second-order). When computed from a cloud (or “plume”) of solutes dissolved in groundwater, moments are commonly used to describe the plume’s total mass (moment of order zero), the location of its centre of mass (first moment), and its dispersion (second moment).

Higher-order moments can also be used to derive quantities that further describe the shape of the solute distribution. These include for example the coefficient of skewness, or degree of asymmetry (third moment), and kurtosis, or relative peakedness or flatness (fourth moment). One must note, however, that statistics of order two or higher are quite sensitive to outlier values [*Press et al.*, 1992]. Caution should therefore be used in the interpretation of these moments, which may not be appropriate for significantly distorted solute plumes.

The theory of spatial moments is summarized below, along with a discussion of their physical interpretation. Several properties can also be derived from the moments to provide additional information about the spatial characteristics of solute plumes. These are described in the following sections along with the moments used in their calculation.

1.1 Theory of spatial moments and derived properties

Moments of a set of values, for instance the solute concentrations measured at each sampling point in an aquifer and interpolated on a regular grid, can be defined [*Press et al.*, 1992] as “the sums of integer powers of the values”. They are in fact the sum of concentrations weighted by the coordinates raised to an integer power. In three-dimensional space, the integral representation of the ijk th spatial moment of the spatial distribution of a solute plume is expressed as [e.g. *Freyberg*, 1986]:

$$M_{ijk}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) x^i y^j z^k dx dy dz \quad (C.1)$$

where n is the effective porosity, $C(x, y, z, t)$ is the concentration field, and x, y , and z are the spatial coordinates. The order of the moment is obtained by summing the indices i, j

and k . The product of the porosity and concentration effectively expresses concentration per unit volume of bulk porous medium rather than per unit volume of water. Spatial moments M_{ijk} only provide a measure of the concentration distribution over the extent of the solute plume itself, since concentration $C(x, y, z, t)$ is equal to zero in those regions of an aquifer where no solute is present. This property is referred to as “plume scale” [Freyberg, 1986]. Moments of order higher than zero are usually normalized by the zero-order moment, and moments of order higher than one are centred on the first.

Table C-1 summarizes the physical interpretation of spatial moments up to the fourth order, in the context of a dataset whose values are comprised of solute concentrations integrated over a given portion of aquifer, or “flow domain”.

Table C-1. Interpretation of spatial moments and their time derivatives.

Order	Interpretation	Time derivative
0	Total mass of dissolved solute	Rate of mass gain/loss
1	Location of centre of mass (centroid)	Velocity of centroid (equivalent to the groundwater velocity in this case)
2	Dispersion (spatial variance) around centroid Orientation (with respect to the longitudinal direction, x) Elongation (aspect ratio of long to short plume axes)	Rate of spreading related to the dispersion coefficient and macrodispersivity
3	Coefficient of skewness (degree of symmetry around centroid, compared to a normal (Gaussian) distribution)	
4	Kurtosis (degree of flatness or peakedness, compared to normal (Gaussian) distribution)	

The equations used to derive spatial moments of zero- to fourth-order, as well as other parameters derived from them, are presented below.

Zero-order order moments: total mass

The zero-order spatial moment is obtained by summing concentration values over the entire flow domain. By setting $i = j = k = 0$, (C.1) reduces to

$$M_{000}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) dx dy dz \quad (C.2)$$

Tracking the zero-order moment over time provides a measure of solute mass conservation within the flow system. A decrease in total mass may indicate, for example,

that a destructive process such as biodegradation or radioactive decay is operating on the solute as it flows through the aquifer, or that the solute plume has migrated past the flow domain boundaries.

In cases where a concentration field is derived from numerical simulations or from images acquired during laboratory flow tank experiments, the zero-order moment can also be used to provide a direct verification of solute mass conservation. For instance, an increasing or decreasing trend in total solute mass may indicate excessive numerical errors, or suggest that the image processing procedure has artificially altered the concentration field.

First-order moments: centroid location and bulk velocity

The first moment in each direction is obtained from (C.2) when $i + j + k = 1$ [Freyberg, 1986].

$$M_{100}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) x dx dy dz \quad (C.3)$$

$$M_{010}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) y dx dy dz$$

$$M_{001}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) z dx dy dz$$

The centre of mass of the plume (x_c, y_c, z_c) can be calculated from the first moment by normalizing (C.3) by the zero-order moment:

$$x_c = M_{100}/M_{000} \quad y_c = M_{010}/M_{000} \quad z_c = M_{001}/M_{000} \quad (C.4)$$

Tracking the location of the centroid over time then yields a plume-scale average velocity vector $\mathbf{v}$:

$$\mathbf{v} = \left[\frac{dx_c}{dt}, \frac{dy_c}{dt}, \frac{dz_c}{dt} \right]^T \quad (C.5)$$

where T is the transpose operator.

As discussed by *Freyberg* [1986], v is equal to the pore water velocity in the case where the groundwater flow field is steady and homogeneous. This is also true for stratified aquifers, when the solute source fully penetrates the flow domain, that is, when the source spans the entire thickness of the aquifer [e.g. *Sudicky*, 1983]. For instantaneous point sources, this equality is valid only for large times, when the vertical dimension of the solute plume is large enough to react to the bulk flow velocity distribution. It is generally assumed [*Freyberg*, 1986] that this relationship also holds true for more complex heterogeneous porous media.

Second-order moments: variance, orientation and elongation

Second order spatial moments are obtained from (C.2) when $i + j + k = 2$. Because these moments are expressed relative to some fixed datum in space, they vary according to their location with respect to this origin. For that reason, it is useful to express second-order moments relative to the plume's centre of mass by:

$$\overline{M}_{ijk}(t) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} n C(x, y, z, t) (x - x_c)^i (y - y_c)^j (z - z_c)^k dx dy dz \quad (C.6)$$

where $i + j + k = 2$; the overbar indicates a central moment. The spatial covariance tensor σ^2 provides an indication of the spread (dispersion) of the solute distribution around the centroid. Its terms are defined by [*Freyberg*, 1986]:

$$\sigma^2 = \begin{bmatrix} \sigma_{xx}^2 & \sigma_{xy}^2 & \sigma_{xz}^2 \\ \sigma_{yx}^2 & \sigma_{yy}^2 & \sigma_{yz}^2 \\ \sigma_{zx}^2 & \sigma_{zy}^2 & \sigma_{zz}^2 \end{bmatrix} \quad (C.7)$$

$$\sigma_{xx}^2 = \overline{M}_{200} / M_{000} \quad \sigma_{yy}^2 = \overline{M}_{020} / M_{000} \quad \sigma_{zz}^2 = \overline{M}_{002} / M_{000}$$

$$\sigma_{xy}^2 = \sigma_{yx}^2 = \overline{M}_{110} / M_{000} \quad \sigma_{xz}^2 = \sigma_{zx}^2 = \overline{M}_{101} / M_{000} \quad \sigma_{yz}^2 = \sigma_{zy}^2 = \overline{M}_{011} / M_{000}$$

The time behaviour of this tensor can be used to derive the hydrodynamic dispersion coefficient $\mathbf{D}$ and macrodispersivity $\mathbf{A}$ tensors that describe the dispersive characteristics

of the plume over time [e.g. *Freyberg, 1986; Farrell et al., 1994a*]. Refer to Appendix E for more details about the calculation and significance of **D** and **A**.

Third-order moments: symmetry (skewness)

Only a few hydrogeological investigations have analyzed spatial moments of order greater than two [e.g. *Gelhar et al., 1979; Naff, 1990; Farrell et al., 1994a*]. Higher-order statistical descriptors can provide information about the spatial distribution of an evolving solute plume. For instance, the coefficient of skewness (derived from the third moment) or kurtosis (determined from the fourth moment) can be used to test the shape of a plume for normality. Monitoring the temporal evolution of these parameters can provide useful information that allows a “real” migrating solute cloud to be characterized, and compared to a hypothetical Gaussian plume, that is, a plume that is normally distributed in space.

The third-order centralized moment is obtained when $i + j + k = 3$ in (C.6). The symmetry of the plume’s distribution in space can be assessed and compared with a normal plume [*Farrell et al., 1994a*] from:

$$SK_x = \frac{\overline{M}_{300}}{M_{000}(\sigma_{xx}^2)^{3/2}} \quad SK_y = \frac{\overline{M}_{030}}{M_{000}(\sigma_{yy}^2)^{3/2}} \quad SK_z = \frac{\overline{M}_{003}}{M_{000}(\sigma_{zz}^2)^{3/2}} \quad (C.8)$$

where SK_x , SK_y , and SK_z represent the coefficients of skewness in the longitudinal (x), transverse (y) and vertical (z) directions, respectively. For a normal distribution, we expect $SK_x = SK_y = SK_z = 0$ [*Press et al., 1992*]. Positive values of skewness indicate a distribution with an asymmetric tail extending towards positive x, y or z, while a tail extending towards the more negative direction of the axes will yield a negative skewness.

Fourth-order moments: flatness (kurtosis)

Similarly, the fourth-order moment can be calculated from (C.6) if $i + j + k = 4$. The peakedness or flatness of the solute distribution within the plume can be described by [*Farrell et al., 1994a*]:

$$\begin{aligned}
KR_x &= \frac{\overline{M}_{400}}{M_{000}(\sigma_{xx}^2)^2} - 3 & KR_y &= \frac{\overline{M}_{040}}{M_{000}(\sigma_{yy}^2)^2} - 3 \\
KR_z &= \frac{\overline{M}_{004}}{M_{000}(\sigma_{zz}^2)^2} - 3
\end{aligned} \tag{C.9}$$

where KR_x , KR_y , and KR_z are defined as the kurtosis along the x , y and z directions, respectively. The -3 terms in (C.12) were inserted so that $KR_x = KR_y = KR_z = 0$ for a normal spatial distribution [Press *et al.*, 1992]. A flatter than normal distribution will have a negative kurtosis (platykurtic), while a positive kurtosis is characteristic of a distribution that is more peaked than normal (leptokurtic).

2 SPATIAL MOMENTS DERIVED FROM IMAGES

In this work, spatial moments were calculated from photographs to monitor the evolution of a cloud of dye migrating through a thin flow tank packed with a porous medium. Appendix A provides a detailed description of the experimental setup.

In the case of a two-dimensional space such as an image, the moments M_{ij} of an object can be easily be derived from the general equation (B.2) by dropping the integral in the third dimension (z). Since concentration values estimated from images are obtained at each picture element (pixel) location, they are already integrated over a small but finite area. The integral notation used up to this point can therefore be dropped in favour of a summation in two dimensions. (B.2) then becomes:

$$M_{ij}(t) = \sum_x \sum_y nC(x, y)x^i y^j \Delta x \Delta y \tag{C.10}$$

where x and y are the coordinates of each pixel in the calibrated image, and $C(x, y)$ is the dye concentration. Pixel dimensions are expressed as Δx and Δy in the horizontal and vertical dimensions, respectively. Although they are always identical on a computer display monitor, it is possible that $\Delta x \neq \Delta y$ if, for example, the image was acquired using a device whose pixels are not square. If such is the case, images will appear to be stretched when viewed on screen. In this work, the aspect ratio of the pixels (7.8 μm wide by 6.8 μm high) was accounted for in all images prior to spatial moment analysis, by

adjusting pixel dimensions to 0.047 by 0.047 cm². This was achieved by changing the original image size (1134×972) to 1134×847 pixels², as described in Appendix B (“Image Processing”), Section 3.4.

Concentration estimates that are obtained from laboratory or field samples, or perhaps from numerical experiments, have units of mass (or moles) per unit volume of water, and are therefore representative of the pore space concentration at the sample location. These values can be interpolated to provide a continuous spatial distribution. Spatial moments are based on bulk coordinates and the concentrations must be multiplied by the porosity n (as in (C.10)) in order to express the concentration as mass (or moles) per unit volume of bulk porous medium. In this study, the calibration process produces pore water concentrations from a form of bulk concentration: the optical density. For homogeneous porous medium experiments, the porosity is constant throughout, and the porosity term in (C.10) is cancelled out when the moments are normalized by M_{00} . Because porosity was determined to be constant for all porous media (bead size classes) used in this work, the same also applies for heterogeneous porous media. In situations where porosity is a function of space, however, it must be accounted for explicitly in the function used to generate the moments, by substituting n with $n(x,y)$ in (C.10).

Pixel intensities (transmittance) must be expressed in units of optical density before they can be related to solute concentrations [e.g. *Schincariol et al.*, 1993]. This implies that each image must be processed and calibrated (to correct defects and convert raw pixel intensities into units of concentration, C) before spatial moments can be calculated and used to describe the solute plume distribution. This procedure is described in detail in Appendix B (“Image Processing”).

Performing moment calculations on an uncalibrated (raw) image would yield erroneous results for two main reasons. First, the non-linear relationship between pixel intensity (transmittance) and concentration would cause moments calculated from raw images to be biased towards one end of the concentration scale. Second, pixel intensities from raw images acquired using the digital camera actually represent “binary transmittance”, which is inversely proportional to light absorption. Low pixel values (dark) thus correspond to high concentration zones because a greater proportion of light

is absorbed by the tracer in those areas, and vice versa. This is because transmittance is highest in areas of low solute concentration, and raw pixel intensities therefore have a high value in low concentration zones. For 8-bit greyscale calibrated images, zero-concentration pixels will therefore have a pixel intensity value approaching 255 (white), while pixel values closer to zero indicate the highest distinguishable solute concentration.

An important source of error in the calculation of the spatial moments stems from random error in the concentration field at some distance away from the plume, where the concentration should in effect be zero. Even if the concentration values are small, the fact that they are located far from the centre of mass induces a large error. As can be seen from the moment generating equation (C.10), the concentrations are weighted by the distance from the centre of mass raised to a power equal to the order of the moment being calculated. Consequently, the concentration measured at pixels located far away from the centroid can influence moment calculations, even though they may be very small. As the pixel location where artificial concentration values are observed increases, their detrimental impact on moment estimation gradually becomes more important. Moreover, this effect is gradually amplified for a given image as the order of the moments increases. This is counter to one of the underlying assumptions that the solute distribution “has a sufficiently strong central tendency” [Press *et al.*, 1992]. The consequence is that results may become inaccurate, for example, by artificially offsetting the location of the centroid, or enhancing the reported value of dispersion. Artificial background noise is an unavoidable consequence of the image processing steps. Consequently, non-zero concentrations are likely to occur across the domain under investigation, which in areas far away from the plume should show no mass. In fact, it is not uncommon to encounter small negative pixel values. Caution should therefore be used when computing spatial moments, and it is often necessary to exclude from the calculations calibrated pixel values lower than a reasonable minimum threshold, C_T .

The cutoff level (lower threshold) is determined for each experiment by the noise level of the image sequence, and is typically set between 1% and 5% of the injection concentration C_0 . For solute concentration distributions resulting from numerical

simulations, this threshold is often set to less than 1% of C_0 . Removal of the artificial mass is done by subtracting the threshold value from the pixel concentrations C_{xy} :

$$C'_{xy} = C_{xy} - C_T \quad C'_{xy} \geq 0 \quad (C.11)$$

The quantity C_T must be selected carefully, as the operation effectively removes artificial background concentration values, while preserving the solute plume's concentration distribution.

2.1 Calculation of two-dimensional spatial moments and related parameters

The zero-order moment is obtained from (C.10) when $i = j = 0$:

$$M_{00} = \sum_x \sum_y n_{xy} C_{xy} \Delta x \Delta y \quad (C.12)$$

The first-order moments in the horizontal (x) and vertical (y) directions are calculated using:

$$M_{10} = \sum_x \sum_y n_{xy} C_{xy} x \Delta x \Delta y \quad M_{01} = \sum_x \sum_y n_{xy} C_{xy} y \Delta x \Delta y \quad (C.13)$$

which can then be normalized to estimate the coordinates of the centroid from:

$$x_c = \frac{M_{10}}{M_{00}} \quad y_c = \frac{M_{01}}{M_{00}} \quad (C.14)$$

The spreading or dispersion of a solute plume in groundwater can be described by the second-order moments M_{20} and M_{02} , using (C.10). These moments provide a measure of solute dispersion in the x and y directions, respectively. However, because the plume migrates and is not stationary in space, moments of second order and higher must be centred on the centroid in order to be useful:

$$\overline{M}_{ij} = \frac{1}{M_{00}} \sum_x \sum_y n_{xy} C_{xy} (x - x_c)^i (y - y_c)^j \Delta x \Delta y \quad i + j \geq 2 \quad (C.15)$$

Moments obtained from (C.14) are referred to as *central moments*, because they are expressed relative to the location of the centre of mass [Awcock and Thomas, 1995].

Central second-order moments are obtained from (C.14) when $i + j = 2$, to provide estimates of the variance in the horizontal ($\overline{M}_{20}$) and vertical ($\overline{M}_{02}$) directions, as well as the covariance ($\overline{M}_{11}$):

$$\begin{aligned}\overline{M}_{20} &= \frac{1}{M_{00}} \sum_x \sum_y n_{xy} C_{xy} (x - x_c)^2 \Delta x \Delta y \\ \overline{M}_{02} &= \frac{1}{M_{00}} \sum_x \sum_y n_{xy} C_{xy} (y - y_c)^2 \Delta x \Delta y \\ \overline{M}_{11} &= \frac{1}{M_{00}} \sum_x \sum_y n_{xy} C_{xy} (x - x_c)(y - y_c) \Delta x \Delta y\end{aligned}\tag{C.16}$$

Note that if the spacing is constant, then Δx and Δy are constants that are cancelled out when normalizing with the zero-order moment. The same is true for the porosity, which is also constant (independent of location in space) for both the homogeneous and heterogeneous cases.

Elongation (or “eccentricity”) ε is derived from the second central moments, and can be used to further characterize the shape of the tracer cloud. It is obtained from [Awcock and Thomas, 1995]:

$$\varepsilon = \frac{(\overline{M}_{20} - \overline{M}_{02})^2 + 4\overline{M}_{11}^2}{M_{00}}\tag{C.17}$$

Similarly, the orientation θ of an object (such as a solute plume) relative to the horizontal axis is defined as the angle of the axis of the minimized moment of inertia (second moment). It is also calculated using the second-order central moments with [Awcock and Thomas, 1995]:

$$\theta = \frac{1}{2} \tan^{-1} \left[\frac{2\overline{M}_{11}}{\overline{M}_{20} - \overline{M}_{02}} \right]\tag{C.18}$$

θ is measured in radians and can be converted to degrees by multiplying it by a factor of $180/\pi$.

Eccentricity and orientation are useful, for example, to investigate the effect on plume evolution of a changing flow direction, as the orientation of the plume's "long" axis is expected to gradually rotate until it parallels the new flow direction. Orientation is expected to be small for compact, pseudo-spherical objects, and get larger as the ratio of maximum to minimum distance vectors between the object's centroid and its boundary increases. Eccentricity will increase as the object becomes increasingly distorted.

Figure C-1 illustrates how the shape of a plume reflects on orientation and eccentricity. The sample plume in Figure C-1 (a) was rotated by -90° (b), -45° (c) and 45° (d). This can be monitored using orientation angles (shown on the figure), while the elongation remains constant. The same plume was then stretched in the horizontal (e) and vertical (f) directions, resulting in an increase or a decrease in the plume's eccentricity values, respectively. Note that for a pseudo-spherical object, the elongation is very small (e.g. Figure C-1 (f)) compared to an oval-shaped object, while the orientation is of little significance because of the absence of a clearly defined elongation.

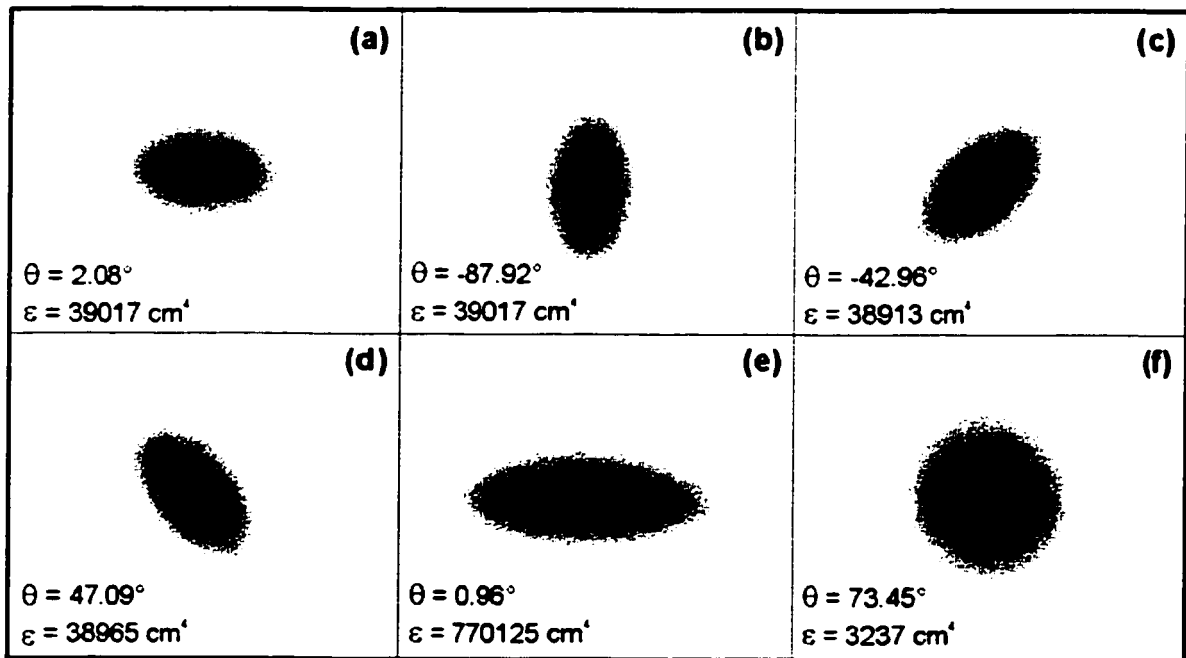


Figure C-1. Illustration of plume orientation and elongation. The sample plume is (a) rotated by -90° (b), -45° (c), and 45° (d), or stretched in the horizontal (e) or vertical (f) direction to illustrate how orientation θ and eccentricity ε are affected. Reported angles are measured clockwise relative to the positive horizontal direction (x-axis).

As before when all three spatial dimensions were considered, third- and fourth-order moments can be computed in two-dimensional space from (C.14) to derive the coefficient of skewness (SK_x and SK_y) and kurtosis (KR_x and KR_y), respectively. (C.8) and (C.9) are then reduced to:

$$SK_x = \frac{\overline{M}_{30}}{M_{00}(\sigma_{xx}^2)^{3/2}} \quad SK_y = \frac{\overline{M}_{03}}{M_{00}(\sigma_{yy}^2)^{3/2}} \quad (C.19)$$

$$KR_x = \frac{\overline{M}_{40}}{M_{00}(\sigma_{xx}^2)^2} - 3 \quad KR_y = \frac{\overline{M}_{04}}{M_{00}(\sigma_{yy}^2)^2} - 3 \quad (C.20)$$

As discussed previously for three-dimensional space in Section 1.1, these parameters will have a value of zero for normal distributions. A positive coefficient of skewness indicates an elongation (asymmetry) in the positive (towards the right) direction. A positive kurtosis indicates a distribution that is more peaked than the Normal, and a negative value is indicative of a distribution that is flatter than the Normal. Kurtosis values between -3 and -1.2 indicate that the plume is multimodal.

In order to calculate spatial moments from calibrated images, a plugin was written and used within the public domain image processing software ImageJ. The following section provides details about the program, as well as on the implementation of this plugin. The source code is also included.

2.2 ImageJ plugin: “Moment Calculator”

ImageJ is a powerful image processing program written in the Java programming language, and available in the public domain. The software and source code are freely available for download¹¹, and is being developed at the National Institute for Mental Health (NIH) by Wayne Rasband. ImageJ was used in this work to process and analyze images, and to calculate spatial moments via the plugin described below. The program, along with a description of its features, documentation, source code, can be found on the Internet at:

¹¹ National Institute of Mental Health, Research Services Branch, Bethesda, MD, <http://rsb.info.nih.gov/ij>

<http://rsb.info.nih.gov/ij/>

Because it was designed with an open architecture, ImageJ provides extensibility through Java plugins. A plugin was developed in order to compute spatial moments of zero- to fourth-order from images. The source code is listed at the end of this appendix, and can be accessed along with the compiled plugin directly from the ImageJ web site at the following location:

<http://rsb.info.nih.gov/ij/plugins/moments.html>

Installation

Download the file “Moment_Calculator.class”. copy it into the “Plugins” folder where ImageJ is installed. and restart the program.

Plugin description

The purpose of this plugin is to calculate the spatial moments up to the fourth-order, as well as derived quantities, from a sequence of greyscale images. In order to be useful, this plugin should be used with images previously standardized or calibrated to a set of known standards present in each image (e.g. pixels expressed in units of optical density, dye concentrations, radioactive isotope standards, etc.). Several curve-fitting methods are available from ImageJ’s ‘Analyze|Calibrate’ menu item, including functions such as: straight line, polynomial (up to 4th order), exponential, log, or Rodbard. The user should experiment to select the calibration function that is most appropriate to their image set. When no internal standards are available, selecting “Uncalibrated OD” from the popup menu causes ImageJ to convert grey values into uncalibrated optical density values using the function:

$$\text{Uncalibrated OD} = \log_{10}(255 / \text{Pixel Value}) \quad (\text{C.21})$$

As a minimum, “raw” images should be inverted if pixels are simply expressed in levels of grey ranging from 0 to 255 (black and white, respectively). This will ensure that the larger pixel values in the image (e.g. the more concentrated portion in a solute plume) correspond to the higher numbers on the measurement scale. In this study, image

sequences were standardized to a set of density standards using the Rodbard function, and pixel values expressed in density units D . This was described in detail in Appendix B.

The moment calculating plugin processes each slice in a sequence (image stack) in two separate passes (iterations). This approach was developed to remove artificial concentrations present as small isolated groups of pixels away from the feature of interest (e.g. solute plume). During the first pass, a large rectangular region of interest (ROI) and relatively high threshold value are specified to obtain a preliminary estimate of first- and second-order moments. These are used to determine the location and extents of the solute plume, which are in turn used to calculate the position and dimensions of the local (oval) ROI used in the second pass. This procedure is illustrated in Figure C-2 for an image from the sequence acquired during the case 7, run 2 experiment.

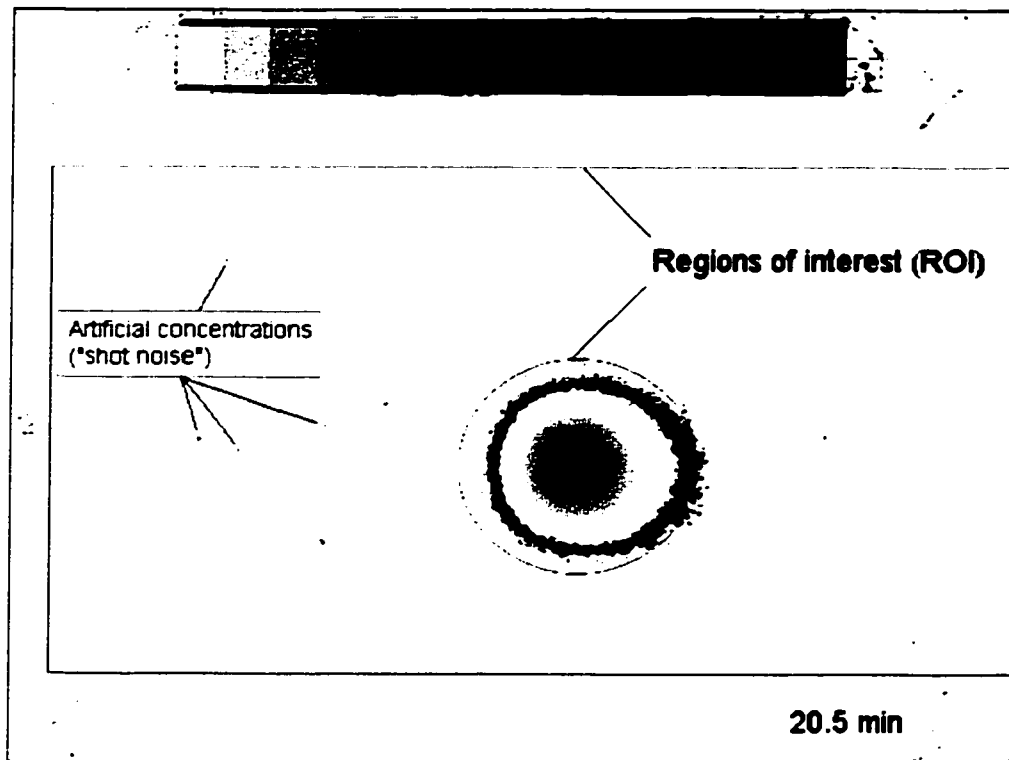


Figure C-2. Sample image (from the case 7, run 2 experiment) illustrating the extents of the regions of interest (ROI) used by the ImageJ plugin "Moment Calculator". Pixels with a standardized density between $D = 0.35$ and $D = 0.50$ are highlighted in black. These correspond to the user-specified "cutoff" threshold values C_2 and C_1 , respectively. The first ROI, a rectangle defined only once by the user and used for all images within a sequence. Pixels within this ROI are used to obtain the preliminary estimates of the location (centroid) and extents (spatial variance) of the solute plume. These then define in a second pass the second (oval) ROI for each image within this sequence. Pixels located outside the local oval ROI are excluded from the final moment calculations, even when their level is above the background threshold (C_2). This procedure filters out unwanted artificial concentrations ("shot noise") present as small groups of pixels. Note that using a single pass with the higher threshold (0.50) would have removed the shot noise, but at the expense of obliterating the details in the diluted portion of the solute plume (the diffuse black "rim", where pixels are in the 0.35-0.50 interval).

The plugin operates as follows:

1. Select a rectangular ROI defining the full extents of the flow domain.
2. From the 'Plugins' menu in ImageJ, select the 'Moment Calculator' plugin
3. In the dialog box (shown in Figure C-3), specify the following:

- a. Specify whether the plugin should process all slices (frames) in a sequence, or only the current image;
- b. Specify which moment derived quantities are to be displayed in the results table;
- c. Enter the “preliminary” parameters (C_1 and N) used to define the dimensions of the local ROI, an oval whose coordinates and extents are computed separately for each slice in an image sequence (see below);
- d. Enter the “final” parameters (C_2 and F) used to calculate the spatial moments from pixels located within the local (oval) ROI. The background (zero concentration) value C_2 is subtracted from the original pixel values. The conversion factor F is then used to adjust pixel values before performing the moment calculations (e.g. unit conversion, linear calibration, etc.). A value of $F = 1$ is used if no conversion is required;
- e. Specify whether a permanent imprint of the local (oval) ROI should be drawn on each image (slice) in a sequence. Note that this permanently alters the images, and therefore should only be used when working from a copy of the image sequence; and
- f. Specify whether pixel values should be converted using a polynomial expression of up to third order prior to moment calculations. The equation, which may be used for example to calibrate standardized pixel values G into concentration units C using a previously determined relationship, is given by:

$$aG^3 + bG^2 + cG + d \quad (C.22)$$

4. Click ‘OK’ to calculate the spatial moments and derived parameters. Results for the quantities selected in step 3.b are displayed in tabular format by ImageJ as they are produced, using a separate row for each image (slice) in a sequence. These results can be saved to disk in ASCII (text) format, and imported into a

spreadsheet application so that they can be plotted and analyzed.


Set Spatial Moments


☒ Process all slices

Check/Uncheck quantities to be displayed in results table.

☒ Image_Name	☒ Mask Size
☒ Total_Mass	☒ X_Centre of Mass
☒ Y_Centre of Mass	☒ X_Variance
☒ Y_Variance	☒ XY_Covariance
☒ X_Skewness	☒ Y_Skewness
☒ X_Kurtosis	☒ Y_Kurtosis
☒ Orientation	☒ Elongation

Enter parameters to define size of oval ROI:

Cutoff1 (C1):

+/- StdDev (N):

Note: A new oval ROI will be defined from the distribution of pixel values contained within the currently selected ROI. The dimensions of this new ROI are set by the standard deviation about the centroid of $[F*(g-C1) > 0]$. The dimensions in X and Y of the new oval ROI are sized using $2*N_xStdDev$ and $2*N_yStdDev$, respectively.

Enter parameters for moment calculations:

Cutoff2 (C2):

Factor (F):

Units:

Note: Statistical moments will be calculated using positive pixel values g (within the oval ROI): $[G = F*(g-C2)] > 0$.

☒ Permanently draw oval ROI (No Undo if checked)

Convert values using polynomial $[C = a*G^3 + b*G^2 + c*G + d]$

a =

b =

c =

d =

Cancel
OK

Figure C-3. Screen capture of the dialog used by the ImageJ plugin "Moment Calculator". The options and parameters that control the behaviour and output preferences of the plugin can be specified via this dialog.

The first ROI (see Figure C-2) is a rectangular area defined by the user prior to running the plugin, and is used throughout the entire image sequence. It should encompass the largest possible area where nothing but the porous medium and tracer are present. For each image in the sequence, pixels located within the rectangular ROI are used to temporarily approximate the coordinates of the plume's centre of mass (x_c and y_c), as well as the spatial variance (σ_{xx}^2 and σ_{yy}^2). The "cutoff" value C_1 is first subtracted from each pixel value G , which is then multiplied by the correction factor F prior to being used to calculate preliminary estimates of the spatial moments (up to second-order). These are in turn used to define the position and dimensions of the local (oval shaped) ROI for each image, as shown in Figure C-2. This local ROI is centred on the preliminary centroid, and extends (on either side of the centroid) up to a distance of $\pm N\sigma_{xx}$ and $\pm N\sigma_{yy}$ in the x and y directions, respectively. Note that these preliminary estimates are calculated for the sole purpose of defining the location and size of the local ROI (oval) for each image. They are discarded and are not used in computing the final spatial moment estimates.

The final estimates of the spatial moments of order up to four and derived quantities are calculated for each image in a sequence as discussed previously, using only those pixels that are located within the local ROI (one per image). This ensures that artificial pixel values (i.e. non-zero concentrations induced by noise) located away from the plume are not included in the moment calculations. As discussed earlier, artificial concentrations can induce large errors in the spatial moment estimates (see §3). The threshold values C_1 and C_2 must therefore be selected carefully. The larger C_1 must be set just high enough to exclude zones of artificial concentrations from the preliminary estimate of the plume's centroid and spatial variance, which are used to define the local (oval) ROI. C_2 is set to a lower value to incorporate the diluted portions of the plume into the "real" moment calculations using pixels located within the local ROI. Pixel values are first adjusted using:

$$G = F (G - C_2) \quad (C.23)$$

and are then used to calculate the final estimates of the spatial moments. Figure C-4 illustrates the importance of selecting an appropriate threshold level. A large value (e.g.

1.00) discards the diluted portion of the plume and makes it seem rather small, while specifying a small threshold incorporates noise as false concentrations.

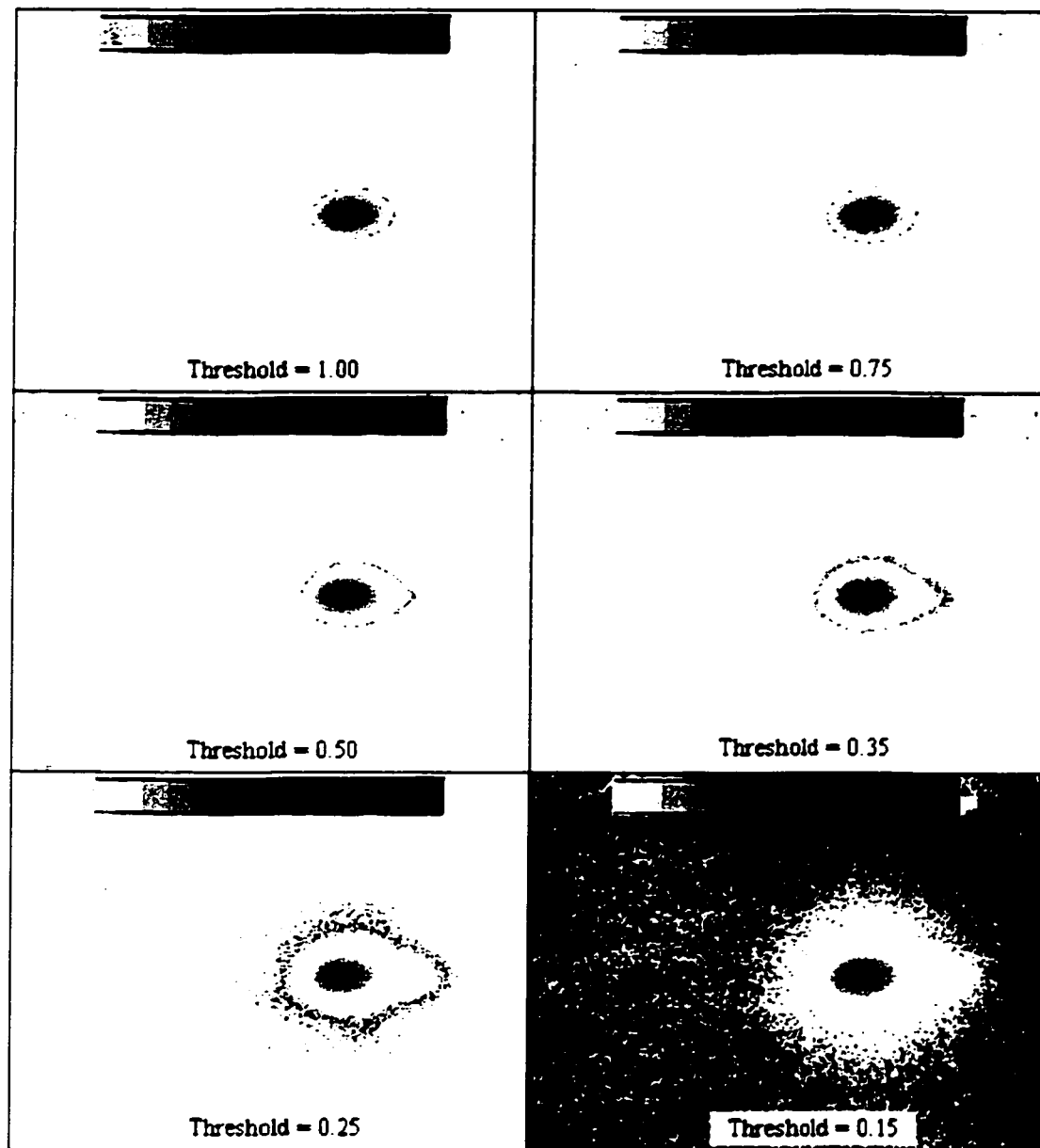


Figure C-4. Impact of threshold selection. A single image from case 2 run 2 is reproduced six times, where pixels whose value corresponds to the arbitrarily selected thresholds shown are highlighted in black.

Plugin source code

Following is a listing of the contents of the source code file "Moment_Calculator.java".

```
import ij.*;
import ij.plugin.filter.*;
import ij.process.*;
import ij.gui.*;
import ij.measure.*;
import ij.text.*;
import java.awt.*;
import java.util.Date;

/* Moment_Calculator plugin

*   @author      Francois Richard
*   @author      University of Ottawa - Earth Sciences
*   @author      richard@science.uottawa.ca

*   @date        12-NOV-2001

This plug-in computes spatial moments up to the 4th order for the
selected (rectangular) ROI, along with some parameters derived from them.
Results are displayed in the main ImageJ window, in addition
to those set for ImageJ's Measure command 'Analyze|Set Measurements'
To export to a text file, use 'File|Save As|Measurements'.
or right-click on the table and select 'Save As'

Note that spatial moments are a very simple and powerful way to describe the
spatial distribution of values, provided they have a sufficiently strong
central tendency, that is, a tendency to cluster around some particular
value. This implies that "background" pixel values are small and that zones
of high concentration (density, etc.) have a high pixel values.

** Interpretation of spatial moments **

* order 0  = TOTAL MASS [units: concentration, density, etc.]
* order 1  = location of CENTRE OF MASS in x and y from 0,0 [units: L]
* order 2  = VARIANCE (spreading) around centroid in x and y [units: L^2]
* order 3  = coeff. of SKEWNESS (symmetry) in x and y [units: n/a]
               --> =0   : SYMMETRIC distribution
               --> <0   : Distribution asymmetric to the LEFT
                       (tail extends left of centre of mass)
               --> >0   : Distribution asymmetric to the RIGHT
                       (tail extends right of centre of mass)
* order 4  = KURTOSIS (flatness) in x and y [units: n/a]
               --> =0   : Gaussian (NORMAL) distribution
               --> <0   : Distribution FLATTER than normal
               --> >0   : Distribution MORE PEAKED than normal
               --> <-1.2: BIMODAL (or multimodal) distribution

** Parameters derived from 2nd moments **
    (from Awcock (1995) "Applied Image Processing")

* ELONGATION (ECCENTRICITY) = Ratio of longest to shortest distance vectors
                           from the object's centroid to its boundaries
* ORIENTATION = For elongated objects, describes the orientation (in
degrees)
                           of the "long" direction with respect to horizontal (x axis)
```

```

*/

public class Moment_Calculator implements PlugInFilter, Measurements {
    ImagePlus imp;
    boolean done;
    ImageWindow win;
    static boolean firstTime = true;
    static boolean processAll = true;
    static boolean show_imageName = true;
    static boolean show_maskSize = true;
    static boolean show_m00 = true;
    static boolean show_xC = true;
    static boolean show_yC = true;
    static boolean show_xxVar = true;
    static boolean show_yyVar = true;
    static boolean show_xyVar = true;
    static boolean show_xSkew = true;
    static boolean show_ySkew = true;
    static boolean show_xKurt = true;
    static boolean show_yKurt = true;
    static boolean show_orientation = true;
    static boolean show_eccentricity = true;
    static boolean draw_ROI = false;
    static double dCutoff1 = 0.50; // cutoff (min. threshold) value - oval ROI
size
    static double dCutoff2 = 0.35; // cutoff (min. threshold) value - moment
calcs
    // static double dFactor = 1.0; // default factor (multiplies pixel values)
    static double dFactor = 0.692; // default factor (multiplies pixel values)
    static String convertedUnits = "N.D.";
    static double dOvalFactor = 4.0; // default factor (determines size of oval
ROI)
    static boolean do_Poly = true;
    static double aPoly3 = 0.06; // factor multiplying G^3 in polynomial equation
    static double bPoly2 = -0.2; // factor multiplying G^3 in polynomial equation
    static double cPoly1 = 1.0; // factor multiplying G^3 in polynomial equation
    static double dPoly0 = 0.0000; // factor multiplying G^3 in polynomial
equation
    public int setup(String arg, ImagePlus imp) {
        if (IJ.versionLessThan("1.23k")) // needs the new PlugInFilter interface
            return DONE;
        if (arg.equals("about")) {showAbout(); return DONE;}
        this.imp = imp;
        if (imp!=null) {
            win = imp.getWindow();
            win.running = true;
        }
        IJ.register(Moment_Calculator.class);
    // return DOES_ALL+DOES_STACKS+NO_CHANGES;
    return DOES_ALL+SUPPORTS_MASKING+ROI_REQUIRED;
    ! // end of 'setup()' method

    public void run(ImageProcessor ip) {
        if (done)
            return;

        int slice,currentSlice,nSlices;
        currentSlice=imp.getCurrentSlice();
        nSlices=imp.getStackSize();
        if(currentSlice != 1){
            IJ.showMessage("Moment Calculator","WARNING...\n"+
                "Processing will start at slice number "+currentSlice+");

```

```

    }

    for(slice=currentSlice;slice<=nSlices;slice++){
        imp.setSlice(slice);

        if (firstTime || Analyzer.getResultsTable().getCounter()==0) {
            if (Analyzer.resetCounter()) {
                setMoments(); // similar to Analyzer|Set Measurements
                if (done)
                    return;
                firstTime = false;
            } else {
                return; // user canceled save changes dialog
            }
        }

        if(dCutoff2>dCutoff1){
            IJ.showMessage("WARNING...", "Cutoff2 larger than Cutoff1\n"+
                "This could lead to funny results...");
        }

        if(!processAll || imp.getStackSize() == 1)nSlices=1; // only current
slice
        int measurements = Analyzer.getMeasurements(); // from Set Measurements
dialog
        Analyzer.setMeasurements(measurements);
        Analyzer a = new Analyzer();
        Calibration cal = imp.getCalibration();
        ImageStatistics stats =
ImageStatistics.getStatistics(ip,measurements,cal);

        // Declare & initialize variables

        double zero = 0.0;
        double m00 = zero;
        double m10 = zero, m01 = zero;
        double m20 = zero, m02 = zero, m11 = zero;
        double m30 = zero, m03 = zero, m21 = zero, m12 = zero;
        double m40 = zero, m04 = zero, m31 = zero, m13 = zero;
        double xC=zero, yC=zero;
        double xxVar = zero, yyVar = zero, xyVar = zero;
        double xSkew = zero, ySkew = zero;
        double xKurt = zero, yKurt = zero;
        double orientation = zero, eccentricity = zero;
        double currentPixel, xCoord, yCoord;

        // Get image and ROI info
        // Note: currently supports rectangular ROIs only

        int width = ip.getWidth();
        int height = ip.getHeight();
        double pw = cal.pixelWidth;
        double ph = cal.pixelHeight;
        boolean isScaled = cal.scaled();
        boolean isCalibrated = cal.calibrated();
        String calUnits = cal.getValueUnit();
        String units = cal.getUnits();
        Roi roi = imp.getRoi();
        Rectangle r = ip.getRoi();
        ip.setCalibrationTable(cal.getCTable());
        String imageName = imp.getTitle()+"."+slice;

```

```

// TEMPORARILY Compute moments of order 0 & 1
// (used only to define location of oval ROI)

for (int y=r.y; y<(r.y+r.height); y++) {
    for (int x=r.x; x<(r.x+r.width); x++) {
        xCoord = (x+0.5)*pw; //this pixel's X calibrated coord. (e.g. cm)
        yCoord = (y+0.5)*ph; //this pixel's Y calibrated coord. (e.g. cm)
        currentPixel=ip.getPixelValue(x,y);
        currentPixel=currentPixel-dCutoff1;
        if (currentPixel < 0) currentPixel = zero; //set negative pixel
values to 0
        currentPixel = dFactor*currentPixel;
/*
        if(do_Poly){ //adjust pixel value using polynomial equation
            currentPixel=aPoly3*Math.pow(currentPixel,3.)+
            bPoly2*Math.pow(currentPixel,2.)+
            cPoly1*currentPixel+
            dPoly0;
        }
*/
/*0*/    m00+=currentPixel;
/*1*/    m10+=currentPixel*xCoord;
        m01+=currentPixel*yCoord;
    }
}

// TEMPORARILY Compute coordinates of centre of mass
// (used only to define horizontal & vertical dims of oval ROI)

if (m00 <= 0.0){
    int lastSlice = slice - 1;
    IJ.showMessage("Moment Calculator","Total mass has reached zero. Plugin
aborted.\n"+
        "Last slice processed: "+lastSlice+".");
    return;
}
xC = m10/m00;
yC = m01/m00;

// Compute origin and dimensions (x & y) of oval ROI.
// * The oval's centre corresponds to the centre of mass (xC,yC)
// * The dimensions of the oval's principal axes correspond to
//   the standard deviation times the user-specified factor
//   (typically equal to 3.0-4.0)

for (int y=r.y; y<(r.y+r.height); y++) {
    for (int x=r.x; x<(r.x+r.width); x++) {
        xCoord = (x+0.5)*pw; //this pixel's X calibrated coord. (e.g. cm)
        yCoord = (y+0.5)*ph; //this pixel's Y calibrated coord. (e.g. cm)
        currentPixel=ip.getPixelValue(x,y);
        currentPixel=currentPixel-dCutoff1;
        if (currentPixel < 0) currentPixel = zero; //set negative pixel
values to 0
        currentPixel = dFactor*currentPixel;
/*
        if(do_Poly){ //adjust pixel value using polynomial equation
            currentPixel=aPoly3*Math.pow(currentPixel,3.)+
            bPoly2*Math.pow(currentPixel,2.)+
            cPoly1*currentPixel+
            dPoly0;
        }
*/

```

```

*/
/*2*/    m20+=currentPixel*(xCoord-xC)*(xCoord-xC);
        m02+=currentPixel*(yCoord-yC)*(yCoord-yC);
    }
}

// Normalize 2nd moments & compute standard deviation around centre of
mass
double xStdDev = Math.pow((m20/m00),0.5);
double yStdDev = Math.pow((m02/m00),0.5);
// Define & create oval ROI
int xOval = (int) ((xC - (dOvalFactor*xStdDev))/pw );
int yOval = (int) ((yC - (dOvalFactor*yStdDev))/ph );
int ovalWidth = (int) ((2.*dOvalFactor*xStdDev)/pw );
int ovalHeight = (int) ((2.*dOvalFactor*yStdDev)/ph );

IJ.makeOval(xOval,yOval,ovalWidth,ovalHeight);
ip = imp.getProcessor();
ip.setCalibrationTable(cal.getCTable());
Rectangle r2 = ip.getRoi();
int[] mask = imp.getMask();

if(draw_ROI){imp.unlock();IJ.run("Draw");win.running=true;}

// Reset moments used above
m00 = zero;
m10 = zero;
m01 = zero;
m20 = zero;
m02 = zero;

// Compute moments of order 0 & 1
int mi,maskSize=0;
double xMin,xMax,yMin,yMax;
int excluded=0;
xMin=(double)((r.x-0.5)*pw);
xMax=(double)((r.x+r.width+0.5)*pw);
yMin=(double)((r.y-0.5)*ph);
yMax=(double)((r.y+r.height+0.5)*ph);
for (int y=r2.y,my=0; y<(r2.y+r2.height); y++,my++) {
    mi = my*r2.width;
    for (int x=r2.x; x<(r2.x+r2.width); x++) {
        if (mask==null || mask[mi++]==ip.BLACK) {
            maskSize++;
            xCoord = (x+0.5)*pw; //this pixel's X calibrated coord. (e.g. cm)
            yCoord = (y+0.5)*ph; //this pixel's Y calibrated coord. (e.g. cm)
            currentPixel=ip.getPixelValue(x,y);
            currentPixel=currentPixel-dCutoff2;    // use new (lower) cutoff
value
            // new Nov12/(excludes pixels extending outside original (rect.)
ROI
            if(xCoord<xMin || xCoord>xMax || yCoord<yMin || yCoord>yMax) {
                currentPixel = zero;
                excluded++;
            }
            if (currentPixel < 0) currentPixel = zero; //gets rid of negative
pixel values
            currentPixel = dFactor*currentPixel;
            if(do_Poly){ //adjust pixel value using polynomial equation
                currentPixel=aPoly3*Math.pow(currentPixel,3.)+
                bPoly2*Math.pow(currentPixel,2.)+

```



```

        cPoly1*currentPixel+
        dPoly0;
    }
/*0*/    m00+=currentPixel;
/*1*/    m10+=currentPixel*xCoord;
        m01+=currentPixel*yCoord;
    }
}

// Compute coordinates of centre of mass

xC = m10/m00;
yC = m01/m00;

// Compute moments of orders 2, 3, 4

for (int y=r2.y,my=0; y<(r2.y+r2.height); y++,my++) {
    mi = my*r2.width;
    for (int x=r2.x; x<(r2.x+r2.width); x++) {
        if (mask==null || mask[mi++]==ip.BLACK) {
            xCoord = (x+0.5)*pw; //this pixel's X calibrated coord. (e.g. cm)
            yCoord = (y+0.5)*ph; //this pixel's Y calibrated coord. (e.g. cm)
            currentPixel=ip.getPixelValue(x,y);
            currentPixel=currentPixel-dCutoff2;
            //new Nov12/01
            if (xCoord<xMin || xCoord>xMax || yCoord<yMin || yCoord>yMax)
currentPixel = zero;
            if (currentPixel < 0) currentPixel = zero; //gets rid of negative
pixel values
            currentPixel = dFactor*currentPixel;
            if (do_Poly){ //adjust pixel value using polynomial equation
                currentPixel=aPoly3*Math.pow(currentPixel,3.)+
                bPoly2*Math.pow(currentPixel,2.)+
                cPoly1*currentPixel+
                dPoly0;
            }
/*2*/    m20+=currentPixel*(xCoord-xC)*(xCoord-xC);
            m02+=currentPixel*(yCoord-yC)*(yCoord-yC);
            m11+=currentPixel*(xCoord-xC)*(yCoord-yC);

/*3*/    m30+=currentPixel*(xCoord-xC)*(xCoord-xC)*(xCoord-xC);
            m03+=currentPixel*(yCoord-yC)*(yCoord-yC)*(yCoord-yC);
            m21+=currentPixel*(xCoord-xC)*(xCoord-xC)*(yCoord-yC);
            m12+=currentPixel*(xCoord-xC)*(yCoord-yC)*(yCoord-yC);

/*4*/    m40+=currentPixel*(xCoord-xC)*(xCoord-xC)*(xCoord-xC)*(xCoord-xC);
            m04+=currentPixel*(yCoord-yC)*(yCoord-yC)*(yCoord-yC)*(yCoord-yC);
            m31+=currentPixel*(xCoord-xC)*(xCoord-xC)*(xCoord-xC)*(yCoord-yC);
            m13+=currentPixel*(xCoord-xC)*(yCoord-yC)*(yCoord-yC)*(yCoord-yC);
        }
    }

// Normalize 2nd moments & compute VARIANCE around centre of mass
xxVar = m20/m00;
yyVar = m02/m00;
xyVar = m11/m00;

// Normalize 3rd moments & compute SKEWNESS (symmetry) around centre of
mass
// source: Farrell et al, 1994, Water Resources Research, 30(11):3213-3223

```

```

xSkew = m30 / (m00 * Math.pow(xxVar, (3.0/2.0)));
ySkew = m03 / (m00 * Math.pow(yyVar, (3.0/2.0)));

// Normalize 4th moments & compute KURTOSIS (peakedness) around centre of
mass
// source: Farrell et al, 1994, Water Resources Research, 30(11):3213-3223
xKurt = m40 / (m00 * Math.pow(xxVar, 2.0)) - 3.0;
yKurt = m04 / (m00 * Math.pow(yyVar, 2.0)) - 3.0;

// Compute Orientation and Eccentricity
// source: Awcock, G.J., 1995, "Applied Image Processing", pp. 162-165
orientation = 0.5*Math.atan2((2.0*m11), (m20-m02));
orientation = orientation*180./Math.PI; //convert from radians to degrees
eccentricity = (Math.pow((m20-m02), 2.0)+(4.0*m11*m11))/m00;

a.saveResults(stats, roi); // store in system results table
ResultsTable rt=Analyzer.getResultsTable(); // get the system results
table

if (show_imageName) rt.addLabel("Image", imageName);
if (show_maskSize) rt.addValue("Mask Size", maskSize);
if (show_maskSize) rt.addValue("Excluded from Mask", excluded);
if (show_m00) rt.addValue("Mass", m00);
if (show_xC) rt.addValue("xC", xC);
if (show_yC) rt.addValue("yC", yC);
if (show_xxVar) rt.addValue("xxVar", xxVar);
if (show_yyVar) rt.addValue("yyVar", yyVar);
if (show_xyVar) rt.addValue("xyVar", xyVar);
if (show_xSkew) rt.addValue("xSkew", xSkew);
if (show_ySkew) rt.addValue("ySkew", ySkew);
if (show_xKurt) rt.addValue("xKurt", xKurt);
if (show_yKurt) rt.addValue("yKurt", yKurt);
if (show_orientation) rt.addValue("Orient.", orientation);
if (show_eccentricity) rt.addValue("Elong.", eccentricity);

int counter = rt.getCounter();
if(counter==1) {
    updateHeadings(rt); // update the worksheet headings
    Date date = new Date(); //get today's date
    IJ.write(" { "-date-" }");
    IJ.write(" Image = "-imageName);
    if(isScaled) {
        IJ.write(" Scaled image [ distance units = "+units+" ].");
    } else {
        IJ.write(" No spatial calibration [results in pixels].");
    }
    if(isCalibrated) {
        if(dFactor == 1.){
            IJ.write(" Calibrated image [ density units = "+calUnits+" ].");
        } else {
            IJ.write(" Calibrated image [ original units (from image info) =
"+calUnits+" ].");
            IJ.write(" ** new units specified by user = "+convertedUnits);
        }
    } else {
        IJ.write(" No density calibration [uncalibrated results].");
    }
    IJ.write(" Cutoff1 (C1) = "+dCutoff1+" (used to define oval ROI)");
    IJ.write(" Cutoff2 (C2) = "+dCutoff2+" (used to calculate moments
within oval ROI g=g-C2>=0)");
    IJ.write(" Factor (F) = "+dFactor+" (used to scale pixel values (g)
using G=F*g)");
}

```

```

IJ.write(" Oval ROI extends +/- "+dOvalFactor+" standard deviations"+
        " from the centre of mass");
if(do_Poly) {
    double test=aPoly3*Math.pow(6.,3.)+
        bPoly2*Math.pow(6.,2.)+
        cPoly1*6.+
        dPoly0;
    IJ.write(" Polynomial conversion used [ C=aG^3+bG^2+cG+d ]:");
    IJ.write("      a="+aPoly3+"; b="+bPoly2+"; c="+cPoly1+"; d="+dPoly0+"
[ G=7 => C="+test+" ]");
} else {
    IJ.write(" Polynomial conversion not used.");
}

IJ.write(rt.getRowAsString(counter-1));

if (!win.running) {
    done = true;
    slice = nSlices;
    IJ.beep();
    IJ.showMessage(" Moment Calculator ", " Operation aborted by user \n"+
        " Stack processing
incomplete");
    IJ.showStatus(" Moment Calculator plugin aborted ...");
}

imp.setRoi(r); //restore initial rectangular ROI

} // end of 'for(slice...' loop
} // end of 'run()' method

// Prompt user to select moments to display & set cutoff value
public void setMoments() {
    GenericDialog gd = new GenericDialog("Set Spatial Moments");
    gd.addCheckbox(" Process all slices ",true);
    gd.addMessage("Check/Uncheck quantities to be displayed in results
table: \n");
    String[] labels = new String[14];
    boolean[] states = new boolean[14];
    labels[0]=" Image_Name "; states[0]=true;
    labels[1]=" Mask_Size "; states[1]=true;
    labels[2]=" Total_Mass "; states[2]=true;
    labels[3]=" X_Centre of Mass "; states[3]=true;
    labels[4]=" Y_Centre of Mass "; states[4]=true;
    labels[5]=" X_Variance "; states[5]=true;
    labels[6]=" Y_Variance "; states[6]=true;
    labels[7]=" XY_Covariance "; states[7]=true;
    labels[8]=" X_Skewness "; states[8]=true;
    labels[9]=" Y_Skewness "; states[9]=true;
    labels[10]=" X_Kurtosis "; states[10]=true;
    labels[11]=" Y_Kurtosis "; states[11]=true;
    labels[12]=" Orientation "; states[12]=true;
    labels[13]=" Elongation "; states[13]=true;
    gd.addCheckboxGroup(7,2,labels,states);
    gd.addMessage("Enter parameters to define size of oval ROI: \n");
    gd.addNumericField(" Cutoff1 (C1): ", dCutoff1, 3);
    gd.addNumericField(" +/- StdDev (N): ", dOvalFactor, 2);
    gd.addMessage("Note: A new oval ROI will be defined from the
distribution\n");
}

```

```

        " of pixel values contained within the currently
selected ROI.\n"+
        " The dimensions of this new ROI are set by the
standard\n"+
        " deviation about the centroid of [ F*(g-C1) > 0 ].\n"+
        " The dimensions in X and Y of the new oval ROI are\n"+
        " sized using 2*N*xStdDev and 2*N*yStdDev,
respectively.");
        gd.addMessage("Enter parameters for moment calculations: \n");
        gd.addNumericField(" Cutoff2 (C2): ", dCutoff2, 3);
        gd.addNumericField(" Factor (F): ", dFactor, 3);
        gd.addStringField("Units:", convertedUnits, 15);
        gd.addMessage("Note: Statistical moments will be calculated using\n"+
        " positive pixel values g (within the oval ROI): [ G =
F*(g-C2) ] > 0.");
        gd.addCheckbox(" Permanently draw oval ROI (No Undo if checked) ",
false);
        gd.addCheckbox(" Convert values using polynomial [ C =
a*G^3+b*G^2+c*G+d ] ", true);
        gd.addNumericField(" a = ", aPoly3, 4);
        gd.addNumericField(" b = ", bPoly2, 4);
        gd.addNumericField(" c = ", cPoly1, 4);
        gd.addNumericField(" d = ", dPoly0, 4);
        gd.showDialog();
        if (gd.wasCanceled()) {
            done = true;
            return;
        }
        processAll = gd.getNextBoolean();
        show_imageName = gd.getNextBoolean();
        show_maskSize = gd.getNextBoolean();
        show_m00 = gd.getNextBoolean();
        show_xC = gd.getNextBoolean();
        show_yC = gd.getNextBoolean();
        show_xxVar = gd.getNextBoolean();
        show_yyVar = gd.getNextBoolean();
        show_xyVar = gd.getNextBoolean();
        show_xSkew = gd.getNextBoolean();
        show_ySkew = gd.getNextBoolean();
        show_xKurt = gd.getNextBoolean();
        show_yKurt = gd.getNextBoolean();
        show_orientation = gd.getNextBoolean();
        show_eccentricity = gd.getNextBoolean();
        dCutoff1 = (double)gd.getNextNumber();
        dOvalFactor = (double)gd.getNextNumber();
        dCutoff2 = (double)gd.getNextNumber();
        dFactor = (double)gd.getNextNumber();
        convertedUnits = gd.getNextString();
        draw_ROI = gd.getNextBoolean();
        dc_Poly = gd.getNextBoolean();
        aPoly3= (double)gd.getNextNumber();
        bPoly2= (double)gd.getNextNumber();
        cPoly1= (double)gd.getNextNumber();
        dPoly0= (double)gd.getNextNumber();
    }

    public void updateHeadings(ResultsTable rt) { // Wayne Rasband
        TextPanel tp = IJ.getTextPanel();
        if (tp==null)
            return;
        String worksheetHeadings = tp.getColumnHeadings();

```

```

        String tableHeadings = rt.getColumnHeadings();
        if (!worksheetHeadings.equals(tableHeadings))
            IJ.setColumnHeadings(tableHeadings);
    } // end of 'updateHeadings' method

    // Create a message about this plugin in 'Help|About Plugins' submenu
    // (must also modify IJ_Props.txt and add it to ij.jar for this to show up)

    void showAbout() {
        IJ.showMessage("About Moment_Calculator...",
            " This plug-in computes spatial moments up to the 4th order\n" +
            " for the selected ROI.\n" +
            " Results are displayed in the main ImageJ window, in addition\n" +
            " to those set for ImageJ's Measure command 'Analyze|Set
Measurements'\n" +
            " To export to a text file, use 'File|Save As|Measurements'.\n" +
            " or right-click on the table and select 'Save As'\n"+
            " \n" +
            " ** Interpretation of spatial moments ** \n" +
            " \n" +
            " * order 0 = TOTAL MASS [units: concentration, density,
etc.]\n"+
            " \n"+
            " * order 1 = location of CENTRE OF MASS in x and y from 0,0
[units: L]\n"+
            " * order 2 = VARIANCE (spreading) around centroid in x and y
[units: L^2]\n"+
            " * order 3 = coeff. of SKEWNESS (symmetry) in x and y [units:
n/a]\n"+
            " --> =0 : SYMMETRIC distribution\n"+
            " --> <0 : Distribution asymmetric to the
LEFT\n"+
            " (tail is to left of centre of
mass)\n"+
            " --> >0 : Distribution asymmetric to the
RIGHT\n"+
            " (tail is to left of centre of
mass)\n"+
            " * order 4 = coeff. of KURTOSIS (flatness) in x and y [units:
n/a]\n"+
            " --> =0 : Gaussian (NORMAL) distribution\n"+
            " --> <0 : Distribution FLATTER than normal\n"+
            " --> >0 : Distribution MORE PEAKED than
normal\n"+
            " --> <-1.2: BIMODAL distribution\n"+
            " \n"+
            " ** Parameters derived from 2nd moments ** (from Awcock (1995) 'Applied
Image Processing')\n"+
            " \n"+
            " * ELONGATION = Ratio of longest to shortest distance vectors\n"+
            " (ECCENTRICITY) from the object's centroid to its boundaries\n"+
            " \n"+
            " * ORIENTATION = For elongated objects, the orientation (in
degrees)\n"+
            " of the 'long' direction with respect to horizontal
(x)\n"+
            " \n");
    } // end of 'showAbout()' method
} // end of 'Moment_Calculator' class

```

APPENDIX D – Macrodispersivity Calculations

*There's no sense in being precise when you
don't even know what you're talking about*

– John von Neumann

Mechanical mixing in a porous medium depends on the velocity with which water flows through the medium and on the ability of the medium to mix water: this property is called macrodispersivity, often expressed as A . As explained in detail in the following section, mechanical mixing, i.e. the coefficient of dispersion, can be calculated directly from the time-rate of change of the spatial variance of the solute plume, and the velocity from the time-rate of change of the position of the plume's centre of mass. Macrodispersivity, on the other hand, must be derived from the coefficient of dispersion and from the velocity.

Since both the coefficient of dispersion and the velocity are based on time derivatives, the method by which the derivatives are estimated are of utmost importance. This appendix describes the methods that were used to estimate the time derivatives, and how the macrodispersivities were calculated. The general formulae used to derive macrodispersivity are presented below, followed by a description of the four calculation methods used in this study. Finally, results from two laboratory experiments are used to derive macrodispersivity estimates using all four methods, and demonstrate the importance of method selection.

1 BACKGROUND

The macroscopic dispersive behaviour of the dissolved tracer can be characterized by calculating the macrodispersivity tensor A_{ij} from [Freyberg, 1986]:

$$A_{ij} = \frac{1}{\|\mathbf{v}\|} D_{ij} \quad (\text{D.1})$$

where $i, j = 1, 2$ is the longitudinal (x) or transverse (y) direction, respectively, D_{ij} is the two-dimensional dispersion coefficient tensor, and $\|\mathbf{v}\|$ represents the magnitude of the solute plume's bulk velocity.

Both D_{ij} and $\|\mathbf{v}\|$ are obtained from the experimental or numerical results. D_{ij} is equal to one half the time rate of change of the spatial covariance tensor σ_{ij}^2 , estimated from the second central moment (see Appendix C), and is given by:

$$D_{ij} = \frac{1}{2} \frac{d\sigma_{ij}^2}{dt} \quad (\text{D.2})$$

Similarly, the components of the velocity vector can be calculated from the change in time of the location of the plume's centre of mass:

$$\bar{v}_i = \frac{d\bar{x}_i}{dt} \quad (\text{D.3})$$

Its magnitude is defined as:

$$\|\mathbf{v}\| = (\bar{v}_i^2 + v_j^2)^{1/2} \quad (\text{D.4})$$

2 CALCULATION METHODS FOR A_{ij}

The estimation of macrodispersivity in (D.1) requires the selection of an “appropriate” time interval to evaluate the time derivatives of $\sigma_{ij}^2(t)$ and $\bar{x}_i(t)$. First, early and late data values should be rejected, because a portion of the solute has either not entered (at early times) or left (at late times) the flow domain, and consequently the spatial moments and derived quantities are incorrect. In particular, the range of values to be included can be selected by monitoring the total mass (moment of order zero), as well as the spatial variance of the plume (second central moments). Once an “acceptable” range is identified (containing N data points), one of several possible methods can be used to estimate the time derivatives that are used to calculate D_{ij} and $\|\mathbf{v}\|$, and thus A_{ij} . Four such methods were identified, including the *Global*, *Stepwise*, *Absolute Stepwise* and *Finite Difference* methods. These

methods are described below, and their application is illustrated using data from selected laboratory experiments.

2.1 “Global” Method

In the *Global* method, all N values within the “acceptable” range are used to produce a single estimate of the time derivatives of $\sigma_{ij}^2(t)$ and $\bar{x}_i(t)$ by linear regression. The small-scale variability in the slope is averaged out by the regression process. The mean squared error on the slope (MSE) provides an estimate of the variability in estimating $\bar{v}_i$ or D_{ij} . It is obtained from:

$$MSE = \frac{\sum (y - \hat{\mu}_{yx})^2}{DF} \quad (D.5)$$

where $\sum (y - \hat{\mu}_{yx})^2$ is the residual sum of squares (SSE), a measure of the error introduced by using the mean $\hat{\mu}_{yx}$ from the linear regression to approximate the measured variable y (e.g. $\bar{v}_i$ or D_{ij}) at a given time t . The MSE has units of (slope) squared, which are not a practical measure of uncertainty. Instead, the positive root of the MSE is used. This quantity is called the standard error of the slope (SE), and it is essentially the standard deviation of the estimated slope.

The main advantage of the *Global* method is that it is quick and simple, as it does not require breaking down the experimental data into sub-intervals for analysis. The procedure is illustrated in Figure D-1 for a laboratory experiment (Case 4, run 1). In Figure D-1(a), $\bar{v}_x$ (shown as a solid line) is estimated from the time-derivative of longitudinal component of the position vector (x_c) using (3.1). This produces a unique estimate of $\bar{v}_x$ that is independent of time (Figure D-1(b)).

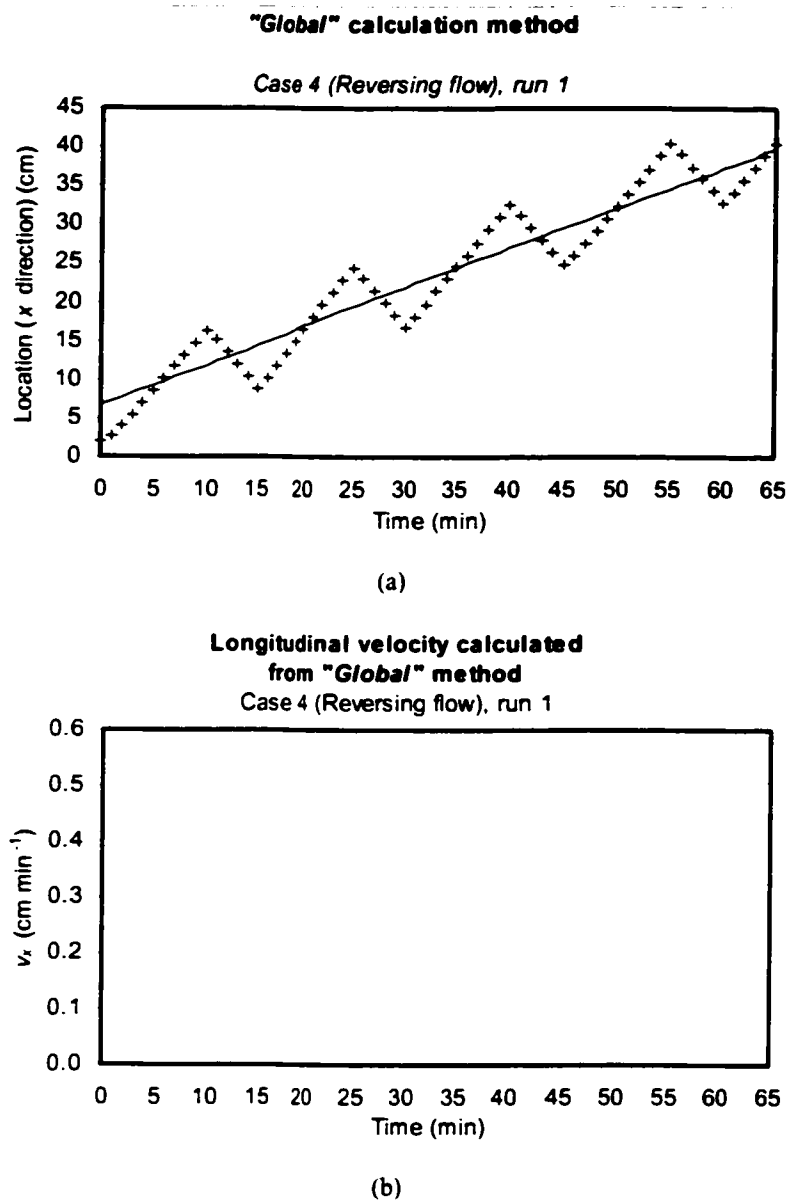


Figure D-1. Schematic example for a reversing flow scenario (case 4, run 1) of the *Global* method for calculating the longitudinal component of velocity, $\bar{v}_x$. The linear regression (solid line) in (a) was derived from the plume's centroid location over time (crosses), using data from the interval $t = 4$ -65 min. The *Global* estimate (b) of velocity ($\bar{v}_x = 0.51 \text{ cm min}^{-1}$) was obtained from the coefficient (slope) of the regression.

2.2 “Stepwise” Method

The purpose of this method is to account for the temporal variations in groundwater flow conditions, in situations where the time series can be divided into M sub-intervals, where M is the number of periods during which flow conditions are held constant. The parameters $\bar{v}_i(m)$ and $D_{ij}(m)$ are then evaluated from linear regressions over each sub-interval, indexed on m . Macrodispersivity can be calculated in two ways. The first, producing an “integrated” estimate of macrodispersivity, is the ratio of the mean D_{ij} value to the mean $\bar{v}_i$ value, where “mean” is understood to be the weighted arithmetic average using the time duration of each sub-interval as the weighting factor. The second method, producing a “time-averaged” macrodispersivity, is the mean of the macrodispersivities estimated over each sub-interval; here again, “mean” is understood to be a time-weighted average.

The time-averaged value of D_{ij} or $\bar{v}_i$ are obtained by averaging the M intermediate estimates, weighted by the duration of the sub-interval $\Delta t(m)$:

$$\langle D_{ij} \rangle = \frac{1}{2} \frac{\sum_{m=1}^M D_{ij}(m) \cdot \Delta t(m)}{\sum_{m=1}^M \Delta t(m)} = \frac{1}{2} \frac{\sum_{m=1}^M \left(\frac{d\sigma_{ij}^2}{dt} \cdot \Delta t \right)_m}{\sum_{m=1}^M \Delta t(m)} \quad (D.6)$$

$$\bar{v}_i = \frac{\sum_{m=1}^M \bar{v}_i(m) \cdot \Delta t(m)}{\sum_{m=1}^M \Delta t(m)} = \frac{\sum_{m=1}^M \left(\frac{d\bar{x}_i}{dt} \cdot \Delta t \right)_m}{\sum_{m=1}^M \Delta t(m)} \quad (D.7)$$

This procedure is illustrated in Figure D-2 for $\bar{v}_x$ using data from the same experiment as before (case 4, run 1). The “integrated” macrodispersivity was estimated by:

$$\bar{A}_{ij} = \frac{\langle D_{ij} \rangle}{\langle \|\mathbf{v}\| \rangle} \quad (D.8)$$

and the “time-averaged” macrodispersivity from the average of the stepwise dispersivities and velocity:

$$\langle A_{ij} \rangle = \sum_{m=1}^M \left[\frac{D_{ij}(m)}{\|\mathbf{v}(m)\|} \cdot \Delta t(m) \right] / \sum_{m=1}^M \Delta t(m) \quad (\text{D.9})$$

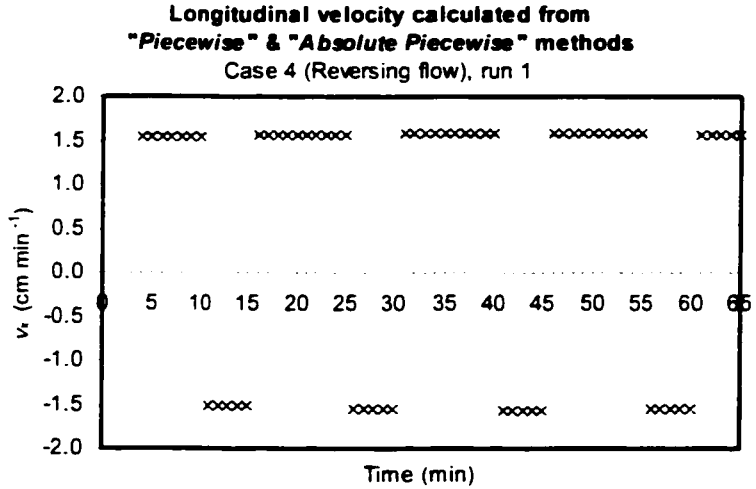
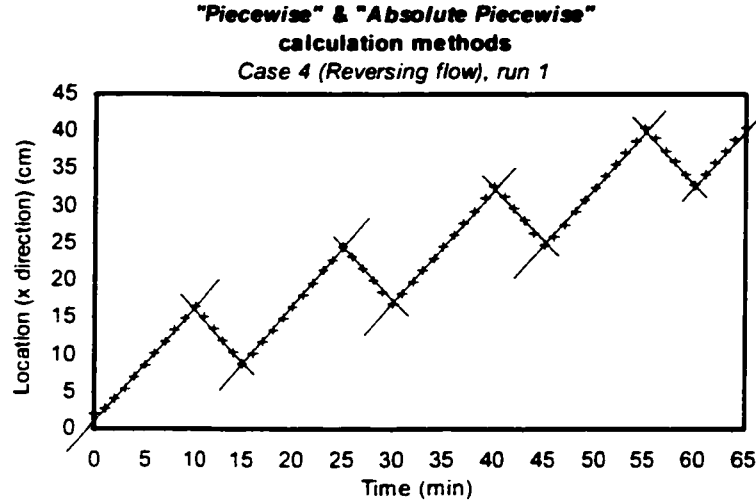


Figure D-2. Schematic example for a reversing flow scenario of the *Stepwise* and *Absolute Stepwise* methods for calculating velocity. The solid lines in (a) show the slope estimates obtained from the nine sub-intervals during which flow conditions were uniform. These slopes, whose values are alternately positive and negative, were weighted by the length of their time sub-intervals, and averaged to calculate the *Stepwise* estimate of $\langle \bar{v}_x \rangle = 0.55 \text{ cm min}^{-1}$ (shown as a dashed line in (b)). A similar approach was used in the *Absolute Stepwise* method (see §2.3), with the exception that the absolute values of the slopes were used in the weighted average, resulting in an estimate $\langle \bar{v}_x \rangle = 1.57 \text{ cm min}^{-1}$, almost three times larger than when using the *Stepwise* method (solid line in (b)).

2.3 “Absolute Stepwise” Method

This method is particularly relevant to cases where the direction of the hydraulic gradient is altered. In such situations, one or both component(s) of the velocity vector can become negative relative to a fixed Cartesian coordinate system, which in the previous method would result in underestimating $\bar{v}_i$. For instance, in “zigzag” cases where bulk groundwater movement periodically changes on either side of the mean gradient direction, the *Global* or *Stepwise* methods will produce an estimate of $\langle \bar{v}_i \rangle$ close to zero because the positive and negative segments are cancelled out in (D.6). But since the solute plume is still experiencing movement along that direction (whether $\bar{v}_i$ is negative or positive), it would appear logical to use the absolute values of each component of the velocity vector in the above summation. This is equivalent to saying that the plume always moves in the positive x and/or y direction, thereby treating the real path as if the plume were actually only migrating along the positive x and y directions.

Accordingly, the data are subdivided into M segments as in the previous method, with the exception that the absolute values of $\bar{v}_i(m)$ are used in the averaged estimate of the bulk velocity, so that (D.6) becomes:

$$\langle \bar{v}_i \rangle = \frac{\sum_{m=1}^M |\bar{v}_i(m)| \cdot \Delta t(m)}{\sum_{m=1}^M \Delta t(m)} = \frac{\sum_{m=1}^M \left(\left| \frac{d\bar{x}_i}{dt} \right| \cdot \Delta t \right)_m}{\sum_{m=1}^M \Delta t(m)} \quad (\text{D.10})$$

The result from (D.10) is shown as a solid line in Figure D-2(b) for a reversing scenario (Case 4, run 1). The “integrated” and “time-averaged” macrodispersivity estimates are evaluated identically as in the *Stepwise* method, using (D.7) and (D.8), respectively (see §2.2).

2.4 “Finite Difference” Method

Calculations are made for each pair of adjacent points, and the individual estimates ($N - 1$ in total) are then averaged using:

$$\overline{D_{ij}} = \frac{1}{2} \frac{\sum_{n=2}^N [\sigma_{ij}^2(n) - \sigma_{ij}^2(n-1)]}{\sum_{n=2}^N [\iota(n) - \iota(n-1)]} = \frac{1}{2} \frac{\sigma_{ij}^2(N) - \sigma_{ij}^2(1)}{\iota(N) - \iota(1)} \quad (\text{D.11})$$

$$\overline{\bar{v}_i} = \frac{\sum_{n=2}^N [\bar{x}_i(n) - \bar{x}_i(n-1)]}{\sum_{n=2}^N [\iota(n) - \iota(n-1)]} = \frac{\bar{x}_i(N) - \bar{x}_i(1)}{\iota(N) - \iota(1)} \quad (\text{D.12})$$

Figure D-3 illustrates this procedure. Similarly, the “integrated” macrodispersivity thus becomes:

$$\overline{A_{ij}} = \frac{1}{2} \frac{[\sigma_{ij}^2(N) - \sigma_{ij}^2(1)]}{\{ [\bar{x}_1(N) - \bar{x}_1(1)]^2 + [\bar{x}_2(N) - \bar{x}_2(1)]^2 \}^{1/2}} \quad (\text{D.13})$$

The “time-averaged” macrodispersivity fully captures the changing trends in each time series, if any, and thus increases the variability in the estimation of A_{ij} . It is given by:

$$\langle A_{ij} \rangle = \frac{1}{2} \frac{\sum_{n=2}^N [\sigma_{ij}^2(n) - \sigma_{ij}^2(n-1)] \cdot \sqrt{[\bar{x}_1(N) - \bar{x}_1(1)]^2 + [\bar{x}_2(N) - \bar{x}_2(1)]^2}}{\sum_{n=2}^N [\iota(n) - \iota(n-1)]} \quad (\text{D.14})$$

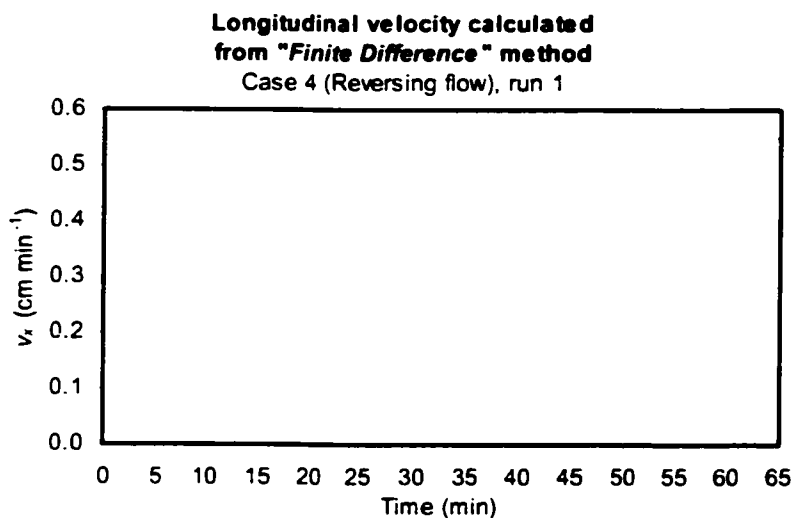
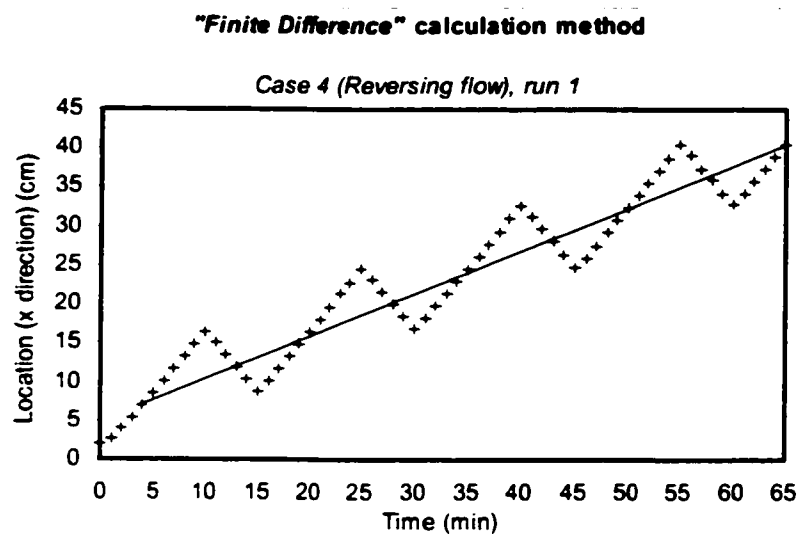
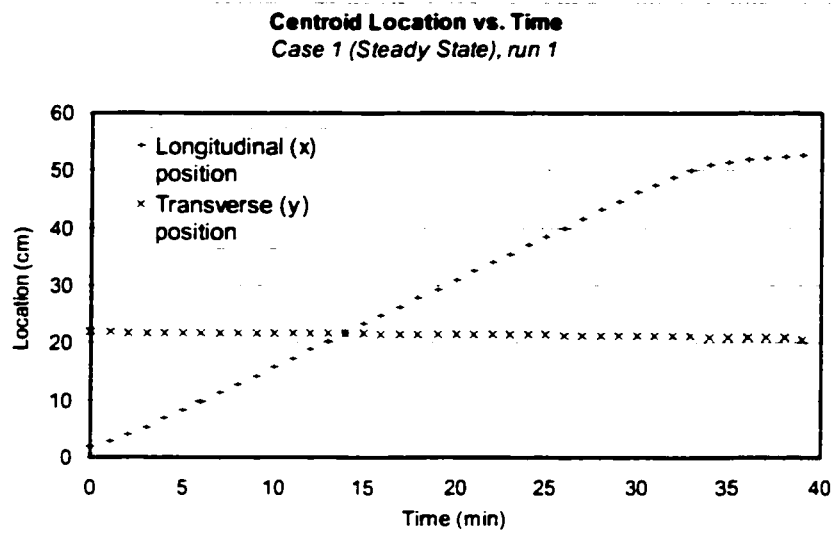


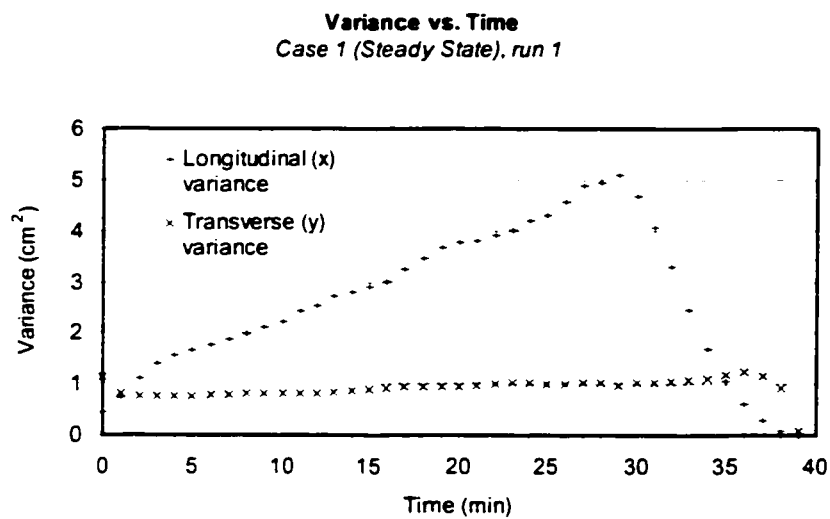
Figure D-3. Schematic example for a reversing flow scenario of the *Finite Difference* method for calculating the longitudinal component of velocity, $\bar{v}_x$. The plume's centroid location over time (crosses) is shown in (a), where the solid line joins the first and last data points used to estimate the mean longitudinal velocity ($t = 4$ and 65 , respectively). The result ($\bar{v}_x = 0.55 \text{ cm min}^{-1}$) is plotted as a solid line in (b).

3 COMPARISON OF THE METHODS

This section evaluates the impact on the calculation of A_{ij} of selecting one of the above methods. As expected, method selection has little impact on A_{ij} for simple flow configurations where flow variations are small or insignificant. For example, consider a scenario of steady state flow in a homogeneous aquifer, where hydraulic gradient direction and magnitude are independent of both space and time (Figure D-4). Macrodispersion A_{ij} and its components were calculated using (D.1)-(D.4) using each of the four methods presented above, using the data comprised in the interval between 3 and 29 minutes. As described earlier, this interval was selected a priori from the examination of the spatial moments, rejecting data at times when it was apparent that a portion of the plume was outside the visible domain. The results for this experiment are presented in Table D-1.



(a)



(b)

Figure D-4. Centroid location (a) and variance (b) versus time for the steady state flow scenario (Case 1, run 1). The time interval used in the calculations is from 3 to 29 minutes, during which the entire solute plume was visible in the images.

Table D-1. Sample experimental data from a steady state flow scenario (Case 1, run 1), where dispersion was calculated from the data in Figure D-4 using each of the four methods discussed in the text. Note that the differences between the methods are insignificant. The two estimates of macrodispersivity (integrated and time-averaged) also produce similar values for a given direction. The overbar and angular brackets are used to differentiate between the integrated and time-averaged methods. See text for details.

Calculation method	N	M	v_x cm min ⁻¹	v_y cm min ⁻¹	$\ v\ $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	27	1	1.515	-0.020	1.515	0.071	0.006	0.047	0.004	-	-
Stepwise	27	1	1.515	-0.020	1.515	0.071	0.006	0.047	0.004	0.047	0.004
Absolute Stepwise	27	1	1.515	0.020	1.515	0.071	0.006	0.047	0.004	0.047	0.004
Finite Difference	-	26	1.517	-0.020	1.517	0.071	0.004	0.047	0.003	0.047	0.003

Because in this scenario flow direction is held constant, there is only a single flow period, and $M = 1$ in the *Stepwise* and *Absolute Stepwise* methods; both methods are thus equivalent to using the *Global* method in this case. The longitudinal and transverse components of the spatial variance in Figure D-4(b) (σ_{11}^2 and σ_{22}^2 , respectively) increase steadily with time, but with minor variability. These slight deviations are incorporated into the calculations by the *Finite Difference* method, resulting in dispersion estimates (D_i and A_i) that deviate slightly from the other three methods.

In the more complex scenarios however, for example when the flow direction is not constant, choosing the appropriate calculation method for A_{ij} is far from trivial. To illustrate this, consider Figure D-5, which shows the evolution in time of the x and y components of the plume centroid and variance for a transient flow scenario. In this case, the homogeneously permeable flow domain was subjected to periodically reversing flow conditions. The evolution of the centroid location in the longitudinal direction (i.e. parallel to flow), $\bar{x}_1$ vs. time (t), shows that the flow regime is made up of a series of repeating cycles. The bulk groundwater movement alternates between the positive x direction for a period of 10 minutes, followed by a “reversing” flow period of 5 minutes during which the flow takes place in the opposite direction ($-x$). In cases such as this, where at least one of the components of the velocity vector $\bar{v}_i$ becomes negative over one or many time interval(s), selection of the appropriate method to compute A_{ij} can lead to significant differences in the results.

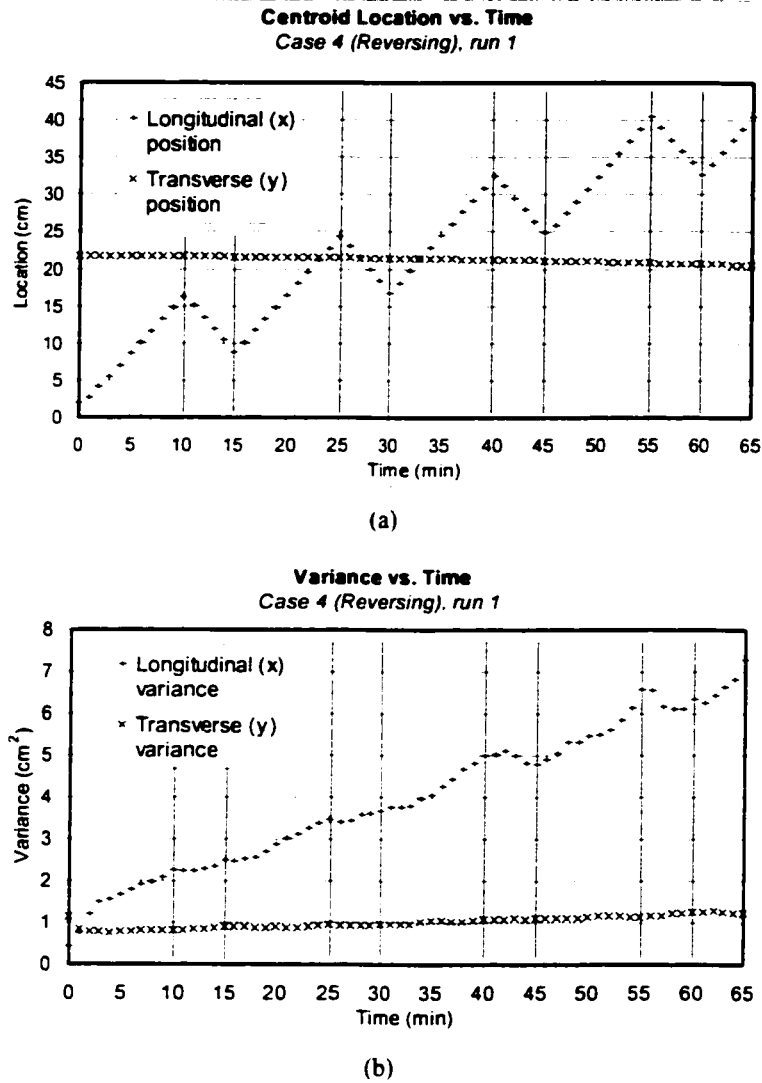


Figure D-5. Centroid location (top) and variance (bottom) versus time for the reversing flow scenarios (Case 4). The time interval used in the calculations is from 4 to 65 minutes, when the full extent of the solute plume was visible in the images. The hydraulic gradient, whose magnitude was held constant ($i = 0.12$), was alternated between the positive and negative longitudinal directions (x and $-x$, respectively). Flow reversals are indicated by the vertical dotted lines.

The experimental data shown in Figure D-5 were used to calculate macrodispersivity using the four methods discussed previously. Because of the transient nature of the flow configuration, method selection can cause very large differences in some of the calculated parameters, as illustrated in Table D-2.

Table D-2. Sample experimental data for a reversing flow scenario (Case 4, run 1). Calculations were performed using the four methods discussed in the text from the data in Figure D-5

Calculation method	<i>N</i>	<i>M</i>	v_x cm min ⁻¹	v_y cm min ⁻¹	$\ \mathbf{v}\ $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
<i>Global</i>	61	1	0.508	-0.019	0.508	0.044	0.004	0.086	0.008	-	-
<i>Stepwise</i>	6	9	0.554	-0.019	0.554	0.045	0.004	0.081	0.007	0.029	0.002
<i>Absolute Stepwise</i>	6	9	1.565	0.019	1.565	0.045	0.004	0.029	0.002	0.029	0.002
<i>Finite Difference</i>	-	60	0.550	-0.019	0.550	0.047	0.004	0.085	0.007	0.028	0.002

Because in this particular case flow velocity was not altered in the transverse (*y*) direction, D_y or $\bar{v}_y$ estimates only show minor differences between calculation methods.

On the other hand, method selection has a considerable impact on macrodispersivity estimates. Results from the *Absolute Stepwise* method particularly contrast with those from the other methods. The results from Table D-2 show $\overline{A_x}$ values ranging between 0.0288 and 0.0859 cm, while $\overline{A_y}$ varies from 0.0024 to 0.0079 cm, depending on the calculation method. These differences are explained in large part by the longitudinal seepage velocity $\bar{v}_x$ estimate, which is much larger when derived using the *Absolute Stepwise* method than for the other methods. Table D-3 shows results from another transient laboratory experiment (Case 2, run 1) where the bulk hydraulic gradient was periodically rotated by $\pm 30^\circ$ in the transverse direction (*y*). As in the previous example, the *Absolute Stepwise* method clearly stands out, with $\bar{v}_y$ – and thus A_y – estimates that contrast with those from the other three methods.

Table D-3. Sample experimental data for flow scenario (Case 2, run 1) where the bulk gradient direction was periodically rotated by $\pm 30^\circ$ relative to the longitudinal (*x*) direction.

Calculation method	<i>N</i>	<i>M</i>	v_x cm min ⁻¹	v_y cm min ⁻¹	$\ \mathbf{v}\ $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
<i>Global</i>	55	1	1.354	-0.013	1.354	0.053	0.013	0.039	0.010	-	-
<i>Stepwise</i>	13	4	1.360	-0.021	1.361	0.053	0.015	0.039	0.011	0.034	0.009
<i>Absolute Stepwise</i>	13	4	1.360	0.754	1.555	0.053	0.015	0.034	0.009	0.034	0.009
<i>Finite Difference</i>	-	54	1.357	-0.022	1.358	0.054	0.013	0.040	0.010	0.035	0.008

Two general observations from the above examples are particularly important to note. First, the “time-averaged” macrodispersivity values $\langle A_i \rangle$ from each of the methods are almost identical to the “integrated” $\bar{A}_i$ obtained when the *Absolute Stepwise* method is used. Second, all parameters calculated using the *Global*, *Stepwise* and *Finite Difference* produce nearly identical results. The *Global* method can thus be considered representative of the *Stepwise* and *Finite Difference* methods, and the latter two methods will not be discussed further to avoid redundancy. Consequently, only results derived from the *Global* and *Absolute Stepwise* methods are shown and discussed in chapters 3 and 4 (from experiments involving homogeneous and heterogeneous porous media, respectively). Similarly, time-averaged macrodispersivity estimates are not included because they do not provide any new information.

4 DETAILED RESULTS

This section presents the macrodispersion estimates calculated using all four methods discussed previously. Laboratory results (presented below, in §4.1) were averaged over η replicates, shown in brackets for each scenario. Results from the individual runs are presented in Appendix F, while the procedure used to average data from multiple runs are provided in §5.

4.1 Laboratory experiments

Table D-4. Experimental results from laboratory experiments, calculated using the four methods discussed in the text (see §2). Data shown for each scenario (case) were averaged over η replicate runs (shown in brackets). Refer to Appendix F to view results from each run individually.

(a) **Case 1 (Steady State, $i = 0.1211$) - All runs ($\eta=6$)**

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	26 1	1.539	-0.017	1.539	0.074	0.005	0.048	0.003	-	-
Stepwise	26 1	1.539	-0.017	1.539	0.074	0.005	0.048	0.003	0.048	0.003
Absolute Stepwise	26 1	1.539	0.017	1.539	0.074	0.005	0.048	0.003	0.048	0.003
Finite Difference	- 25	1.541	-0.017	1.541	0.074	0.004	0.048	0.003	0.048	0.003

(b) **Case 2 (Zig-Zag 30°, $i = 0.1211$) - All runs ($\eta=3$)**

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	42 1	1.365	-0.017	1.365	0.055	0.012	0.040	0.009	-	-
Stepwise	10 4	1.372	-0.039	1.372	0.055	0.012	0.040	0.009	0.035	0.008
Absolute Stepwise	10 4	1.372	0.749	1.563	0.055	0.012	0.035	0.008	0.035	0.008
Finite Difference	- 41	1.370	-0.039	1.370	0.057	0.011	0.041	0.008	0.036	0.007

(c) **Case 3 (Zig-Zag 60°, $i = 0.1211$) - All runs ($\eta=2$)**

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	49 1	0.800	-0.019	0.800	0.026	0.022	0.033	0.028	-	-
Stepwise	8 6	0.804	0.008	0.805	0.027	0.022	0.033	0.027	0.017	0.014
Absolute Stepwise	8 6	0.804	1.312	1.539	0.027	0.022	0.017	0.014	0.017	0.014
Finite Difference	- 48	0.804	0.007	0.804	0.028	0.022	0.035	0.027	0.018	0.014

(d) **Case 4 (Reversing, $i = 0.1211$) - All runs ($\eta=2$)**

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	62 1	0.500	-0.018	0.500	0.042	0.004	0.084	0.009	-	-
Stepwise	7 9	0.565	-0.017	0.565	0.047	0.004	0.082	0.007	0.030	0.002
Absolute Stepwise	7 9	1.536	0.017	1.536	0.047	0.004	0.030	0.002	0.030	0.002
Finite Difference	- 61	0.561	-0.017	0.561	0.048	0.004	0.085	0.007	0.030	0.002

Table D-4 (continued)

Experimental results from laboratory experiments, calculated using the four methods discussed in the text (see §2). Data shown for each scenario (case) were averaged over η replicate runs (shown in brackets). Refer to Appendix F to view results from each run individually.

(e) Case 5 (Steady State, $i = 0.2174$) - All runs ($\eta=2$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ v $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	30 1	2.596	-0.013	2.596	0.120	0.005	0.046	0.002	-	-
Stepwise	30 1	2.596	-0.013	2.596	0.120	0.005	0.046	0.002	0.046	0.002
Absolute Stepwise	30 1	2.596	0.013	2.596	0.120	0.005	0.046	0.002	0.046	0.002
Finite Difference	- 29	2.598	-0.014	2.598	0.126	0.006	0.048	0.002	0.048	0.002

(f) Case 6 (Zig-Zag 30°, $i = 0.2174$) - All runs ($\eta=2$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ v $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	36 1	2.210	0.106	2.213	0.082	0.016	0.037	0.007	-	-
Stepwise	7 5	2.206	0.148	2.211	0.089	0.014	0.040	0.006	0.035	0.005
Absolute Stepwise	7 5	2.206	1.207	2.515	0.089	0.014	0.035	0.005	0.035	0.005
Finite Difference	- 35	2.207	0.177	2.215	0.083	0.016	0.037	0.007	0.033	0.006

(g) Case 7 (Zig-Zag 60°, $i = 0.2174$) - All runs ($\eta=2$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ v $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	59 1	1.314	0.080	1.316	0.044	0.036	0.034	0.027	-	-
Stepwise	8 7	1.321	0.031	1.322	0.044	0.035	0.033	0.026	0.017	0.014
Absolute Stepwise	8 7	1.321	2.139	2.514	0.044	0.035	0.017	0.014	0.017	0.014
Finite Difference	- 58	1.317	0.051	1.318	0.044	0.035	0.034	0.027	0.018	0.014

(h) Case 8 (Reversing, $i = 0.2174$) - All runs ($\eta=2$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ v $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	78 1	0.858	-0.010	0.858	0.069	0.005	0.081	0.006	-	-
Stepwise	7 11	0.971	-0.009	0.971	0.078	0.004	0.080	0.004	0.030	0.002
Absolute Stepwise	7 11	2.517	0.012	2.518	0.078	0.004	0.031	0.002	0.030	0.002
Finite Difference	- 77	0.960	-0.009	0.960	0.076	0.004	0.079	0.004	0.020	0.002

4.2 Numerical simulations

Table D-5. Experimental results from numerical simulations for each scenario (case), calculated using the four methods discussed in the text (see §2).

(a) Case 1 (Steady State, $i = 0.1211$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	28 1	1.585	0.000	1.585	0.076	0.003	-0.048	0.002	-	-
Stepwise	28 1	1.585	0.000	1.585	0.076	0.003	0.048	0.002	0.048	0.002
Absolute Stepwise	28 1	1.585	0.000	1.585	0.076	0.003	0.048	0.002	0.048	0.002
Finite Difference	- 27	1.575	0.000	1.575	0.075	0.002	0.048	0.001	0.048	0.001

(b) Case 2 (Zig-Zag 30°, $i = 0.1211$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	32 1	1.378	-0.008	1.378	0.062	0.021	0.045	0.015	-	-
Stepwise	7 5	1.384	-0.024	1.384	0.065	0.020	0.047	0.015	0.041	0.013
Absolute Stepwise	7 5	1.384	0.739	1.569	0.065	0.020	0.041	0.013	0.041	0.013
Finite Difference	- 31	1.384	-0.025	1.384	0.064	0.019	0.047	0.014	0.041	0.012

(c) Case 3 (Zig-Zag 60°, $i = 0.1211$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	53 1	0.798	-0.001	0.798	0.029	0.056	0.037	0.070	-	-
Stepwise	8 7	0.821	0.001	0.821	0.032	0.054	0.039	0.066	0.020	0.034
Absolute Stepwise	8 7	0.821	1.327	1.561	0.032	0.054	0.021	0.035	0.020	0.034
Finite Difference	- 52	0.822	0.001	0.822	0.032	0.054	0.039	0.066	0.020	0.034

(d) Case 4 (Reversing, $i = 0.1211$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	66 1	0.522	0.000	0.522	0.073	0.004	0.140	0.008	-	-
Stepwise	8 9	0.607	0.000	0.607	0.074	0.004	0.121	0.006	0.047	0.002
Absolute Stepwise	8 9	1.585	0.000	1.585	0.074	0.004	0.047	0.002	0.047	0.002
Finite Difference	- 65	0.607	0.000	0.607	0.074	0.004	0.122	0.006	0.047	0.002

Table D-5 (continued)

Experimental results from numerical simulations for each scenario (case), calculated using the four methods discussed in the text (see §2).

(e) Case 5 (Steady State, $i = 0.2174$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	16 1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	-	-
Stepwise	16 1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	0.042	0.005
Absolute Stepwise	16 1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	0.042	0.005
Finite Difference	- 15	2.884	0.000	2.884	0.122	0.014	0.042	0.005	0.042	0.005

(f) Case 6 (Zig-Zag 30°, $i = 0.2174$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	21 1	2.474	0.118	2.476	0.099	0.045	0.040	0.018	-	-
Stepwise	4 6	2.481	0.185	2.488	0.106	0.043	0.042	0.017	0.037	0.015
Absolute Stepwise	4 6	2.481	1.428	2.862	0.106	0.043	0.037	0.015	0.037	0.015
Finite Difference	- 20	2.474	0.108	2.477	0.096	0.048	0.039	0.019	0.033	0.017

(g) Case 7 (Zig-Zag 60°, $i = 0.2174$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	33 1	1.404	0.084	1.406	0.058	0.093	0.041	0.066	-	-
Stepwise	5 8	1.417	0.005	1.417	0.059	0.095	0.041	0.067	0.021	0.033
Absolute Stepwise	5 8	1.417	2.442	2.824	0.059	0.095	0.021	0.034	0.021	0.033
Finite Difference	- 32	1.418	0.005	1.418	0.058	0.095	0.041	0.067	0.020	0.033

(h) Case 8 (Reversing, $i = 0.2174$)

Calculation method	NM	v_x cm min ⁻¹	v_y cm min ⁻¹	$ \mathbf{v} $ cm min ⁻¹	$\langle D_x \rangle$ cm ² min ⁻¹	$\langle D_y \rangle$ cm ² min ⁻¹	$\overline{A_x}$ cm	$\overline{A_y}$ cm	$\langle A_x \rangle$ cm	$\langle A_y \rangle$ cm
Global	81 1	0.949	0.000	0.949	0.099	0.019	0.104	0.020	-	-
Stepwise	8 11	1.083	0.000	1.083	0.099	0.019	0.091	0.017	0.034	0.007
Absolute Stepwise	8 11	2.885	0.001	2.885	0.099	0.019	0.034	0.007	0.034	0.007
Finite Difference	- 80	1.083	0.000	1.083	0.098	0.019	0.091	0.017	0.000	0.000

5 AVERAGING REPLICATE RUNS AND ESTIMATING UNCERTAINTY

When multiple runs were available for a given scenario, as was the case for laboratory experiments from Chapter 3, the experimental evidence and the associated uncertainty for each run were averaged over all replicate runs η . This section discusses the procedure used to average parameters and to combine precision errors for results calculated using the *Global* and *Absolute Stepwise* methods (§5.1 and §5.2, respectively).

5.1 Global method

As described in §2.1, the experimental error produced by the estimation of the time derivatives of $\bar{x}_i(t)$ and $\sigma_{ij}^2(t)$ was measured using the mean squared error (*MSE*) resulting from the regression estimates (D.5). All calculations and averages were calculated with the *MSE*, but the results are reported as the more relevant standard error (*SE*), which is the square root of the *MSE*. This procedure is used because the *SE* is a biased estimator but the *MSE* is not. The first two rows in Table D-6 show the formulae used to calculate the means over η runs of $\bar{v}_i$ and D_i and their respective uncertainties ((D.17) to (D.22)). The uncertainty for both averaged parameters can be expressed by the weighted average of each run's *MSE*, where each run is weighted by the degrees of freedom *DF*. Because two parameters are estimated by the linear regressions (the slope and the intercept on the ordinate axis), two degrees of freedom are lost in the operation, and $DF = n - 2$, where n represents the number of points in the time interval selected for the regression. The equations used to calculate the variances are given in Table D-6, as $s_{\bar{v}_i}^2$ (D.17) and $s_{D_i}^2$ (D.20). Alternatively, the variability of a given parameter can be expressed relative to the mean by calculating the coefficient of variation *CV* using (D.18) or (D.21)).

For each parameter derived from two or more averaged quantities – each incorporating a degree of uncertainty – the combined error can be calculated using the *superposition-of-errors* theorem [Leaver and Thomas, 1974]. The formulae used to estimate the mean over η runs of $\|\mathbf{v}\|$ and A_i are given in Table D-6 by (D.22) and (D.25), respectively. The uncertainty associated with these derived parameters is also summarized in Table D-6 in

(D.23) and (D.26). These equations were obtained by deriving the following equation for each parameter u [adapted from *Leaver and Thomas, 1974*]:

$$s_u^2 = (\partial u / \partial x)^2 s_x^2 + (\partial u / \partial y)^2 s_y^2 + 2(\partial u / \partial x)(\partial u / \partial y) s_{xy} \quad (\text{D.15})$$

Note that to calculate $s_{\mathcal{A}}^2$ (D.26), the covariance $s_{D,|\mathbf{v}|}$ was neglected since $\|\mathbf{v}\|$ was constant for a given scenario, obtained from $\bar{v}_1$ and $\bar{v}_2$, which were already averaged over η runs using (D.16).

Table D-6. Formulae used to estimate the mean over η runs of certain parameters, as well as the associated uncertainty, which can be expressed by the variance s^2 or by the coefficient of variation CV . See below for a description of the symbols used.

Parameter (averaged over η runs)	Uncertainty (error)
$\bar{v}_i = \left\{ \sum_{k=1}^{\eta} \left[\left(\frac{d\bar{x}_i}{dt} \right) \cdot n \right]_k \right\} / \sum_{k=1}^{\eta} n_k \quad (\text{D.16})$	$s_{\bar{v}_i}^2 = \left[\sum_{k=1}^{\eta} \left[(MSE_{\bar{v}_i}) \cdot DF \right]_k \right] / \sum_{k=1}^{\eta} DF_k \quad (\text{D.17})$
	$CV = \frac{s_{\bar{v}_i}}{\bar{v}_i} \quad (\text{D.18})$
$D_i = \left\{ \sum_{k=1}^{\eta} \left[\left(\frac{1}{2} \frac{d\sigma_i^2}{dt} \right) \cdot n \right]_k \right\} / \sum_{k=1}^{\eta} n_k \quad (\text{D.19})$	$s_{D_i}^2 = \left[\sum_{k=1}^{\eta} \left[\left(\frac{1}{2} MSE_{D_i} \right) \cdot DF \right]_k \right] / \sum_{k=1}^{\eta} DF_k \quad (\text{D.20})$
	$CV = \frac{s_{D_i}}{D_i} \quad (\text{D.21})$
	$s_{\ \mathbf{v}\ }^2 = \left(\frac{\partial \ \mathbf{v}\ }{\partial \bar{v}_1} \right)^2 s_{\bar{v}_1}^2 + \left(\frac{\partial \ \mathbf{v}\ }{\partial \bar{v}_2} \right)^2 s_{\bar{v}_2}^2 + 2 \left(\frac{\partial \ \mathbf{v}\ }{\partial \bar{v}_1} \right) \left(\frac{\partial \ \mathbf{v}\ }{\partial \bar{v}_2} \right) s_{\bar{v}_1 \bar{v}_2}$ $= \left(\frac{\bar{v}_1}{\sqrt{\bar{v}_1^2 + \bar{v}_2^2}} \right)^2 s_{\bar{v}_1}^2 + \left(\frac{\bar{v}_2}{\sqrt{\bar{v}_1^2 + \bar{v}_2^2}} \right)^2 s_{\bar{v}_2}^2 +$ $2 \left(\frac{\bar{v}_1}{\sqrt{\bar{v}_1^2 + \bar{v}_2^2}} \right) \left(\frac{\bar{v}_2}{\sqrt{\bar{v}_1^2 + \bar{v}_2^2}} \right) s_{\bar{v}_1 \bar{v}_2}$ $= \frac{\bar{v}_1^2}{\bar{v}_1^2 + \bar{v}_2^2} s_{\bar{v}_1}^2 + \frac{\bar{v}_2^2}{\bar{v}_1^2 + \bar{v}_2^2} s_{\bar{v}_2}^2 + 2 \frac{\bar{v}_1 \bar{v}_2}{\bar{v}_1^2 + \bar{v}_2^2} s_{\bar{v}_1 \bar{v}_2} \quad (\text{D.22})$
	$CV = \frac{s_{\ \mathbf{v}\ }}{\ \mathbf{v}\ } \quad (\text{D.23})$
	$s_{A_i}^2 = \left(\frac{\partial A_i}{\partial D_i} \right)^2 s_{D_i}^2 + \left(\frac{\partial A_i}{\partial \ \mathbf{v}\ } \right)^2 s_{\ \mathbf{v}\ }^2 + 2 \left(\frac{\partial A_i}{\partial D_i} \right) \left(\frac{\partial A_i}{\partial \ \mathbf{v}\ } \right) s_{D_i \ \mathbf{v}\ }$ $= \left(\frac{1}{\ \mathbf{v}\ } \right)^2 s_{D_i}^2 + \left(\frac{D_i}{-\ \mathbf{v}\ ^2} \right)^2 s_{\ \mathbf{v}\ }^2 + 2 \left(\frac{1}{\ \mathbf{v}\ } \right) \left(\frac{D_i}{-\ \mathbf{v}\ ^2} \right) s_{D_i \ \mathbf{v}\ }$ $= \frac{1}{\ \mathbf{v}\ ^2} s_{D_i}^2 + \frac{D_i^2}{\ \mathbf{v}\ ^4} s_{\ \mathbf{v}\ }^2 - 2 \frac{D_i}{\ \mathbf{v}\ ^3} s_{D_i \ \mathbf{v}\ } \quad (\text{D.24})$
$A_i = \frac{D_i}{\ \mathbf{v}\ } \quad (\text{D.25})$	$CV = \frac{s_{A_i}}{A_i} \quad (\text{D.26})$
	$CV = \frac{s_{A_i}}{A_i} \quad (\text{D.27})$

- i : Principal components of Cartesian coordinate system (1: longitudinal (x); 2: transverse (y))
 η : No. of replicate runs for a given scenario (case)
 k : Index on run number for a given scenario ($k = 1, \eta$)
 n_k : No. of points used in linear regression to approximate the time rate of change of a parameter
 u : Mean of parameter u , averaged over η runs
 s_u^2 : Variance of mean estimate u , where $u = \bar{v}_i, \|\mathbf{v}\|, D_i$, or A_i)
 MSE_k : Mean squared error from linear regression estimate
 DF_k : Degrees of freedom of means derived from linear regressions ($DF_k = n_k - 2$)
 $\bar{v}_i$: Principal components of seepage velocity vector $\mathbf{v}$, averaged over η runs
 $\bar{x}_i$: Principal components of the position vector $\mathbf{x}$
 $\|\mathbf{v}\|$: Velocity vector magnitude
 D_i : Principal components of the coefficient of dispersion, averaged over η runs
 σ_i^2 : Principal components of the spatial variance
 A_i : Principal components of the macrodispersivity tensor, averaged over η runs

5.2 Absolute Stepwise method

The transient scenarios considered in this study were made of a series of sub-intervals where flow conditions remained unchanged over pre-determined periods. Because changes in flow direction occurred almost instantaneously, the transient experiments can be thought of as a series of steady state experiments of shorter duration; the concentration distributions at the end of a given interval are used as initial conditions for the next period. This is sometimes referred to as “quasi steady state” in the context of numerical studies of groundwater contamination problems [e.g. *Goode and Konikow*, 1990]. A rigorous estimation of the uncertainty associated with data averaged over multiple runs first requires one estimate of the regression error (MSE) for as each sub-interval in a given experiment. Each interval m has $DF(m) = n(m) - 2$ degrees of freedom. The total error for the complete experiment results from the uncertainty introduced by M linear regressions. It is obtained from:

$$\langle MSE \rangle = \frac{1}{M} \sum_{i=1}^M MSE(m) \quad (D.28)$$

The parameters calculated using the *Absolute Stepwise* method and the associated uncertainty estimates can be averaged over all runs η using (D.17) and (D.20) from Table D-6, after replacing MSE by $\langle MSE \rangle$ for each run k .

Rather than adopting this approach – which would have necessitated about 350 linear regressions – an alternative technique was developed that allowed the standard error associated with $\bar{v}_i$ and D_i in transient runs to be approximated using the standard error from steady state runs of corresponding hydraulic gradient magnitude. As noted above, the transient experiments were made up of a series of uniform flow conditions similar to those encountered in the steady state scenarios (case 1 and case 5). The standard error for these simple cases (obtained from single slope) was estimated using the *Global* method by linear regression. The number of points included in the regression for each run varied from $n = 25$ to 27. The results, calculated as outlined in the previous section, are summarized for both steady state scenarios in Table D-7.

Table D-7. Standard errors on the mean velocity and on the mean coefficient of dispersion for the steady state scenarios, averaged over η runs from equations in Table D-6. These estimates, obtained using the *Global* method, were used in the approximation of the standard errors on means calculated from the *Absolute Stepwise* method. See text for details.

	Case 1 ($\bar{n} = 26$) (Steady, $i = 0.1211$)	Case 5 ($\bar{n} = 29$) (Steady, $i = 0.2174$)
$S_{v_i}^{Global}$	0.00166	0.00218
$S_{v_i}^{Global}$	0.00064	0.00143
$S_{D_i}^{Global}$	0.00162	0.00237
$S_{D_i}^{Global}$	0.00026	0.00030

The uncertainty on parameters calculated using the *Absolute Stepwise* method and averaged over several runs is given by:

$$S_u^{Absolute Stepwise} = S_u^{Global} \cdot \frac{\sqrt{\bar{n}}^{Global}}{\sqrt{\bar{\phi}}^{Absolute Stepwise}} \quad (D.29)$$

APPENDIX E – Reynolds Number

1 BACKGROUND

Darcy's Law describes the motion of a fluid through porous media, and is only valid for flow velocities that are neither too small nor too large. The generalized form of fluid flow through permeable sediments is given by [Freeze and Cherry, 1979]:

$$v = -K \left(\frac{\partial h}{\partial l} \right)^m \quad (E.1)$$

where v is the specific discharge (volumetric flow rate per unit area of porous medium) [$L_w^3 \cdot L_{PM}^2 \cdot T^{-1}$], K is the hydraulic conductivity [$L_w^3 \cdot L_{PM}^{-1} \cdot T^{-1}$], and $\partial h / \partial l$ is the hydraulic gradient [$L_w \cdot L_{PM}^{-1}$]. Laminar flow only occurs when $m = 1$, in which case (E.1) is linear and corresponds to the classical form of Darcy's Law. Conversely, when flow rates are either below or above the lower and upper limits, $m \neq 1$ and (E.1) then becomes non-linear; as a result, the flow equation is no longer called Darcy's Law [Freeze and Cherry, 1979]. This may occur in geological materials where permeability is extremely low (e.g. in compacted clay) and/or hydraulic gradients are extremely small, and groundwater is essentially immobile. This situation is uncommon, however, and is not applicable to the experimental conditions encountered throughout this work. On the other hand, there are some realistic hydrogeological conditions where groundwater flow rates are so fast that flow is no longer laminar, but rather becomes turbulent. This situation can arise when v is very large, such as in aquifers hosted in karstic limestone or dolomite. Although the range of validity of the flow law may seem exceedingly restrictive, it is in fact applicable the vast majority of conditions encountered in natural aquifers. This is particularly true for unconsolidated granular materials.

Darcy's Law is applicable to situations where the ratio of inertial to viscous forces does not exceed a certain threshold value [Bear, 1972]. This dimensionless quantity, also known as Reynolds number (Re), is estimated using [Freeze and Cherry, 1979]:

$$Re = \frac{\rho v d}{\mu} \quad (E.2)$$

where ρ is the fluid density [$M_w \cdot L_w^{-3}$], μ is the kinematic fluid viscosity [$M_w \cdot L^{-1} \cdot T^{-1}$], v is the specific discharge [$L_w^3 \cdot L_{PM}^{-2} \cdot T^{-1}$] and d is a representative length dimension [L].

Although it may take many forms (e.g. mean grain diameter, pore dimension, square root of permeability, etc.), d is commonly taken as the mean grain diameter.

Bear [1972], following a review of the experimental evidence, suggested that "Darcy's Law is valid as long as the Reynolds number based on average grain diameter does not exceed some value between 1 and 10" (p. 126). Flow rates in loose sediments almost never exceed the upper limit of validity of Darcy's Law [Freeze and Cherry, 1979].

In the laboratory, however, it is technically possible to impose on the physical model hydraulic gradients that may be so high as to invalidate Darcy's Law. It is therefore useful to calculate a conservative estimate of Re using the highest expected value of v that one expects from the controlled experimental conditions. This exercise provides reassurance that the flow rates are within the limits of Darcian flow. The following section provides the details of the Re calculations.

2 CALCULATIONS OF RE AND RELATED QUANTITIES

The upper limit of Re was calculated to provide an estimate of the highest acceptable hydraulic gradient, given the conditions encountered in the experiments described in this work. By defining the threshold between laminar and turbulent flow, Re thus defines the upper end of the range of validity of Darcy's Law. Calculations were performed using conservative (i.e. worse-case scenario) estimates of the parameters used in (E.2). The details are provided in this section.

2.1 Upper limit of Darcian flow rate

At 20°C, $\rho = 0.9982 \text{ g cm}^{-3}$ and $\mu = 0.0101 \text{ g cm}^{-1} \text{ s}^{-1}$ [van der Leeden *et al.*, 1990]. By re-arranging (E.2) and using the maximum value of $\text{Re} = 10$, the specific discharge corresponding to the highest limit of validity for Darcy's Law (v_{upper}) is obtained from:

$$v_{\text{upper}} = \frac{10 \cdot (0.0101 \text{ g cm}^{-1} \text{ s}^{-1})}{(0.9982 \text{ g cm}^{-3})(0.05 \text{ cm})} = 2.02 \text{ cm s}^{-1}$$

This value can be used to verify that flow rates imposed on the system by a set of experimental conditions do not exceed the range of validity of Darcy's Law, that is, to ensure that $v \leq v_{\text{upper}}$.

2.2 Experimental flow rates

The highest values of specific discharge were expected in areas where the coarsest glass beads were used (Class 1, $d_{50} = 0.05 \text{ cm}$). The hydraulic conductivity for this bead class was estimated from the Kozeny-Carman equation [e.g. Bear, 1972, Eq. 5.10.18], and measured using a constant head permeameter, giving $K = 0.200 \text{ cm s}^{-1}$ and $K = 0.182 \text{ cm s}^{-1}$, respectively. The physical properties of the nine classes of beads used in the laboratory experiments are summarized in Appendix A (Table A-1). During the course of this study, the flow system was subjected to a maximum hydraulic gradient equal to

$$i = \Delta h / \Delta L = (22.6 \text{ cm} - 9.15 \text{ cm}) / 60.50 \text{ cm} = 0.22$$

where Δh represents the change in hydraulic head between the highest hydraulic heads (e.g. in channels 9 and 10) and the lowest hydraulic heads (channels 3 and 4); ΔL represents the separation distance between ports 9-10 and ports 3-4. Refer to Appendix A (§1.3) for a description of the configuration of the inlet/outlet ports. The maximum specific discharge through these beads was:

$$v_{\text{max}} = -K i = -(0.2 \text{ cm s}^{-1}) (-0.22) = 0.044 \text{ cm s}^{-1}$$

It is clear that $v_{\max}$ is significantly smaller than v_{upper} , and we can therefore conclude that flow in this system is laminar, and Darcy's Law can be used to describe flow through the porous media.

3 SICHARDT'S EMPIRICAL FORMULA FOR BORDERLINE GRADIENT

An alternative method available to verify that flow conditions are within Darcy's Law's validity range is Sichardt's empirical formula [*de Marsily*, 1986]:

$$i_{\max} = \frac{\sqrt{K}}{15} \quad (\text{E.3})$$

where $i_{\max}$ is the borderline (maximum) gradient, and K is expressed in units of m day^{-1} .

Using a value of $K = (0.2 \text{ cm s}^{-1}) (0.01 \text{ m cm}^{-1}) (86400 \text{ s day}^{-1}) = 172.8 \text{ m day}^{-1}$, we obtain $i_{\max} = 0.88$ from (E.3), which is well above the largest gradient used during the course of the laboratory experiments ($i = 0.22$).

4 CONCLUSIONS

Using two calculation methods, namely the Reynolds Number and Sichardt's empirical formula for borderline gradient, it was possible to verify that the hydraulic gradients – and resulting average flow rates – encountered throughout this work are well below the threshold values defined as the upper acceptable limits of validity of Darcy's Law.

Appendix F – Wall Effects

The discussion of the laboratory results suggested that solute transport inside the flow cell is significantly influenced by wall effects. The ability to reverse the hydraulic gradients provided a unique opportunity to observe this phenomenon. The temporal evolution of the quantities derived from spatial moments was discussed in Chapter 3 for experiments conducted in a homogeneous porous medium. Wall effects were interpreted as being responsible for a number of unusual observations, in particular about the total mass, spatial variance, coefficient of skewness, and kurtosis.

A brief description of this phenomenon is presented below, followed by a discussion of the impacts of wall effects on the advection and dispersion of a dissolved solute plume migrating through a laboratory flow cell packed with porous media.

1 BACKGROUND

The wall effect is a disturbance in the flow field caused by the way rounded beads tend to self-organize along the walls of the flow cell, only making point contact with the flat surface: the influence of the wall decreases as the distance away from it increases and the arrangement becomes more random [Benenati and Brosilow, 1962]. A small fraction of the dissolved tracer along the top and bottom of the flow domain travels faster than the bulk of the solute plume. This is due to a very thin zone of increased permeability caused by the increased porosity at the interface between the porous medium and the flow cell's smooth walls. Benenati and Brosilow [1962] used an empirical approach to study the variations in void space as a function of distance from the walls of a container packed with a porous medium composed of beads of uniform dimensions. They concluded that the void fraction V_v / V_t (ratio of the volume of voids V_v over total volume V_t), varies from one at the container walls to approximately 0.38 at a distance of less than 4.5 bead

diameters. Note that when the bead diameter is very small relative to the porous medium thickness, the integrated void fraction is reduced from one at the walls to under 0.4 at a distance of about 1.5 bead diameters in the bed of beads. Given the uniform diameter of the beads used here (0.34 mm), the zone affected by the wall effects was on the order of 0.5 mm at each wall (1.0 mm in total, or about 3% of the total thickness of the porous medium).

2 EVIDENCE

As light travels through the porous medium, it is partially absorbed by the tracer (dye) in proportion to both the concentration and volume of the solution. The integrated view across the full thickness of the flow cell in the images does not allow the distinction to be made between increased porosity (which results in an increased dye volume), or increased dye concentration. However, the importance of the wall effects on solute migration can be assessed indirectly by observing results from the reversing flow scenarios. Recall from the laboratory results presented previously that when the mean hydraulic gradient was reversed, the dissolved solute plume was shown to behave differently than under steady state flow conditions. Some of the observations discussed in previous sections are thought to result from wall effects. In particular, the following were observed during flow reversal intervals (see Chapter 3 for details):

1. Higher than expected maximum concentration C_{max}
2. Slower than expected plume spreading rate (i.e. the slope of the spatial variance vs. time becomes less pronounced, or even negative);
3. Decreasing coefficient of skewness, indicating that the plume asymmetry which developed during forward flow intervals is partially eliminated by reversing the hydraulic gradient direction; and
4. Increasing kurtosis as the concentration distribution becomes more peaked.

In order to illustrate how the evolution of these laboratory-derived quantities can be explained by the wall effects, it is convenient to conceptualize the flow domain as being composed of two distinct porous media. As a result, the dissolved plume can be viewed

as a combination of two distinct plumes superimposed in the vertical direction. The “real” plume travels inside the central portion of the flow cell at the same velocity as the groundwater, where flow and transport are unaffected by the wall effects. In the second zone is the “fast” plume area, much thinner, located at the interfaces between the walls of the flow cell and the porous medium, where permeability is higher due to wall effects. In this study, the concentration distributions are estimated from transmitted-light images, which provide a view of the solute plume integrated over the entire thickness of the flow cell. It is therefore impossible to distinguish between the individual contributions of the “real” and “fast” portions of the combined plume. To illustrate this, consider the hypothetical concentration profiles shown in Figure F-1(a) and Figure F-1(b) for scenarios of steady state and a reversing flow conditions, respectively.

Three profiles are shown for each time step in Figure F-1, including the “real” and the “fast” plumes, as well as their sum, the “composite” plume. When the plume is injected at time t_0 , all three profiles are located at the injection point ($x = 0$). The “real” plume (shown by the dashed profiles) migrates at the same velocity as the mean groundwater flow and spreads symmetrically due to hydrodynamic dispersion. After a period of forward flow (t_1), the centroid (peak) of the “fast” plume has travelled a greater distance and contributes in developing the asymmetry towards the right of the “composite” plume. If flow is held steady from then on (Figure F-1(a)), this trend continues throughout the experiment, the macrodispersivity of the composite plume becomes faster than that of the real plume. Moreover, skewness gradually becomes more positive as the plume becomes increasingly distorted towards the right, while at the same time becoming increasingly flatter as the plume spreads out (the kurtosis decreases). Conversely, when the direction of the hydraulic gradient is reversed some time after t_1 , the “fast” plume begins migrating in the opposite direction faster than the “real” plume. While both will continue experiencing the same amount of spreading as if they had continued travelling to the right, the relative superposition of the two plumes produces a “composite” plume that looks quite different (see Figure F-1(b)). Because the centroids of the “real” and “fast” plumes are aligned at time t_2 , the “composite” result is a plume that has a higher peak

(C_{max}), is more symmetrical, more peaked and somewhat less dispersed than in the steady state scenario.

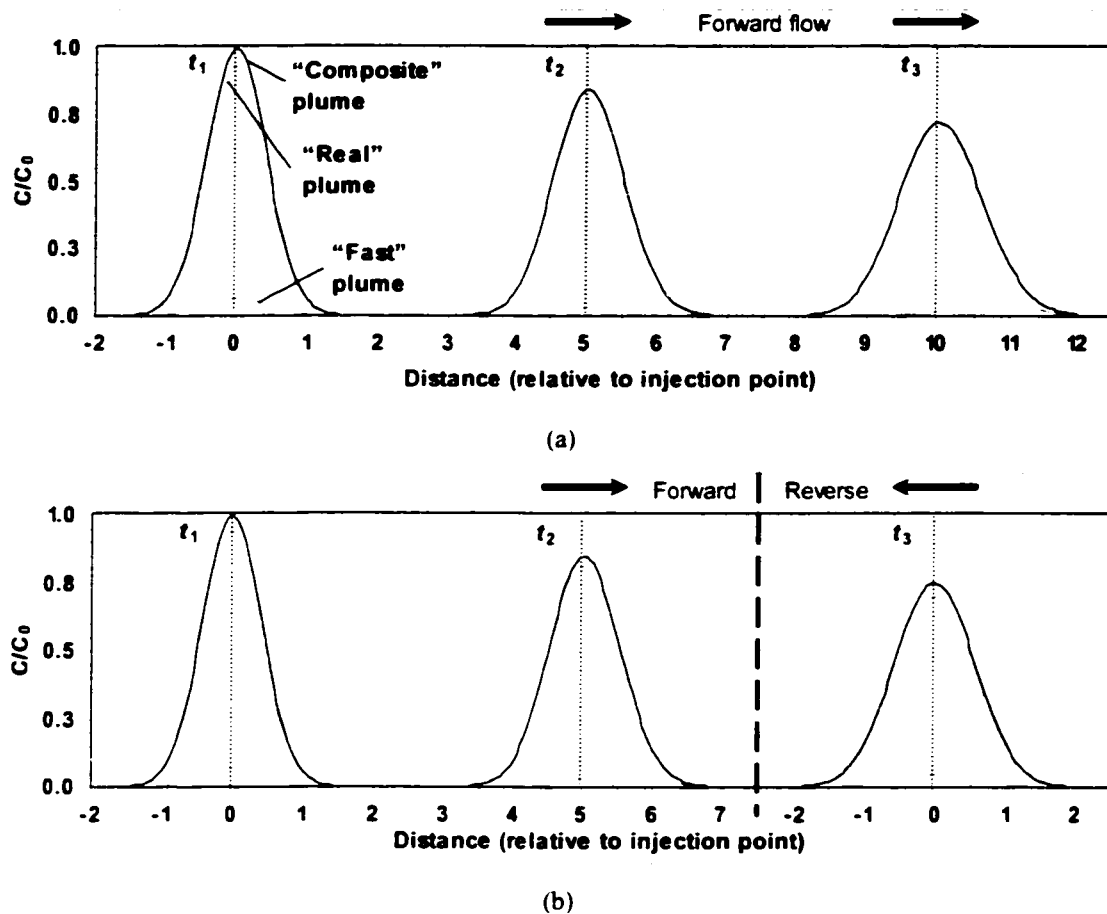


Figure F-1. Schematic diagram illustrating the wall effects observed from laboratory results. The concentration distributions show the longitudinal evolution of a solute plume subjected to steady state (a), or reversing (b) flow conditions. The vertical dotted lines correspond to the predicted centroid location calculated from the groundwater seepage velocity and porosity. To visualize the wall effect, the injected "composite" plume is separated into two "sub-plumes". The first (dashed lines) is the "real" plume that migrates at the same velocity as the mean groundwater flow. The second is a thin "fast" plume, affected by the wall effects whose velocity is higher than that of the "real" plume.

APPENDIX G – Experimental Results

1 GENERAL DESCRIPTION

Experimental results from all runs are presented below. These are presented sequentially in the following order:

1. Homogeneous porous medium experiments (discussed in Chapter 3):
 - a. Laboratory results
 - b. Numerical results
2. Heterogeneous porous medium experiments (discussed in Chapter 4):
 - a. Laboratory results
 - b. Numerical results

Results from each experiment are shown on a separate page, including:

1. Six graphs where the following quantities, derived from spatial moments up to the fourth order (see Appendix C for details) are plotted against time:
 - a. Total mass (M vs. t)
 - b. Centroid location (x_c vs. t and y_c vs. t)
 - c. Spatial variance (σ_{xx} vs. t , σ_{yy} vs. t and σ_{xy} vs. t)
 - d. Orientation and elongation (M vs. t)
 - e. Coefficient of skewness (SK vs. t)
 - f. Kurtosis (KU vs. t)
2. The quantities derived from the temporal evolution of the first- and second-order spatial moments, used to estimate macrodispersivity A_{ij} . These are shown in the table at the bottom of each page as obtained using each of the four

calculation methods discussed in Appendix D. These include the *Global*, *Stepwise*, *Absolute Stepwise* and *Finite Difference* methods, labelled 1 through 4, respectively. The parameters shown for each run are:

- a. N : Total number of data points in the “acceptable” time interval
- b. M : Number of time sub-intervals used by each method
- c. $\bar{v}_x$ and $\bar{v}_y$: Velocity in each direction of the plume centroid
(longitudinal and transverse components, respectively, derived from the plots of x_c vs. t and y_c vs. t .)
- d. $||\mathbf{v}||$: Magnitude of the velocity vector
- e. $\langle D_{xx} \rangle$ and $\langle D_{yy} \rangle$: “Time-averaged” coefficient of dispersion
(longitudinal and transverse estimates, respectively)
- f. $\bar{A}_{xx}$ and $\bar{A}_{yy}$: “Integrated” macrodispersivity (longitudinal and transverse components, respectively)
- g. $\langle A_{xx} \rangle$ and $\langle A_{yy} \rangle$: “Time-averaged” macrodispersivity (longitudinal and transverse components, respectively)

2 RESULTS FROM CHAPTER 3 – HOMOGENEOUS POROUS MEDIUM

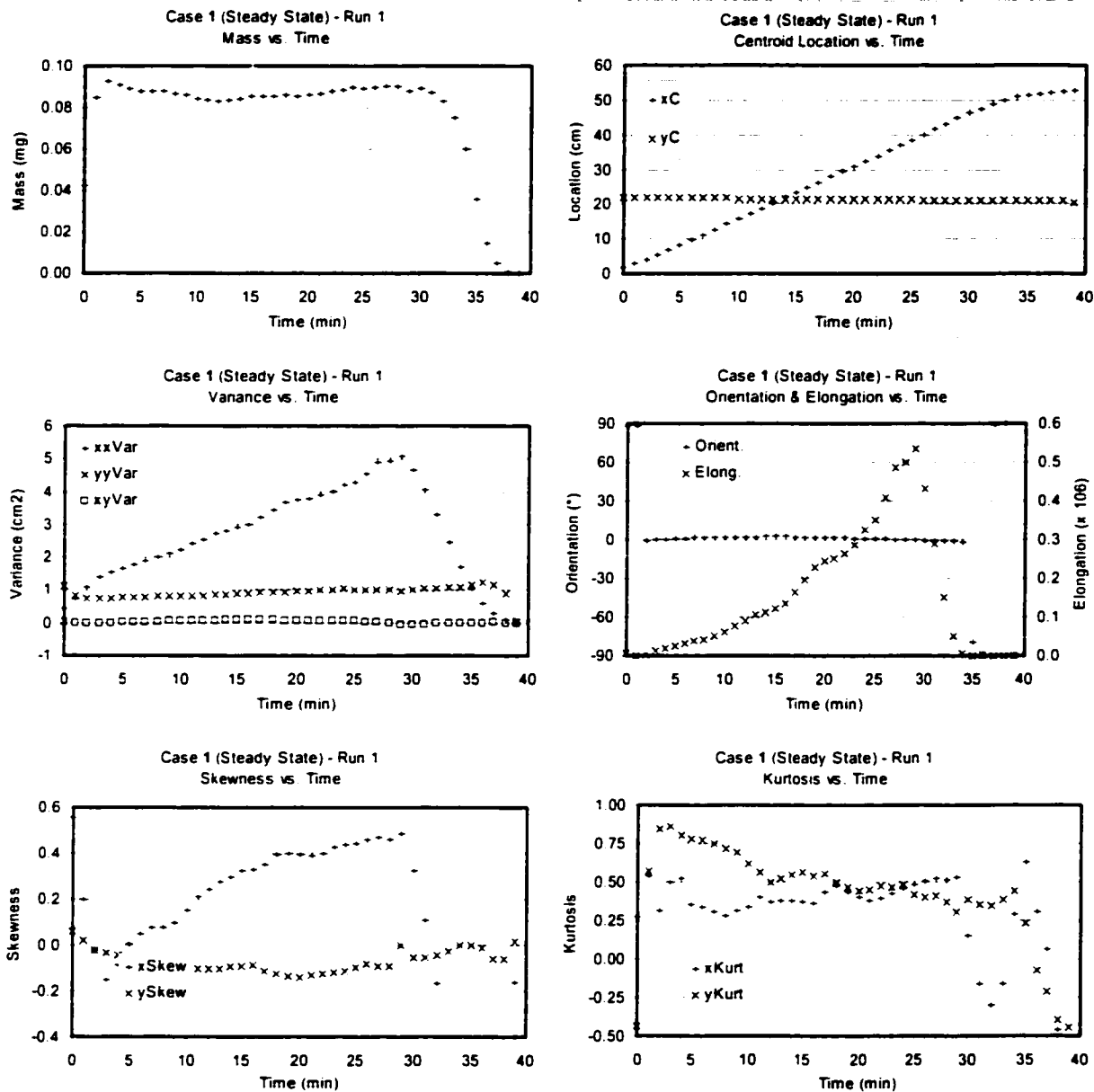
2.1 Laboratory experiments

The following results obtained from the laboratory experiments performed using a homogeneous porous medium (discussed in Chapter 3) are shown below (η represents the number of replicate runs, each shown on a separate page):

1. Hydraulic gradient $i = 0.1211$:
 - a. Case 1: Steady state scenario ($\eta = 6$)
 - b. Case 2: Zigzag 30° scenario ($\eta = 3$)
 - c. Case 3: Zigzag 60° scenario ($\eta = 2$)
 - d. Case 4: Reversing scenario ($\eta = 2$)
2. Hydraulic gradient $i = 0.2174$:
 - a. Case 5: Steady state scenario ($\eta = 2$)
 - b. Case 6: Zigzag 30° scenario ($\eta = 2$)
 - c. Case 7: Zigzag 60° scenario ($\eta = 2$)
 - d. Case 8: Reversing scenario ($\eta = 2$)

Homogeneous Medium - Laboratory Results

Case 1, run 1 (Steady State, $i = 0.1211$)



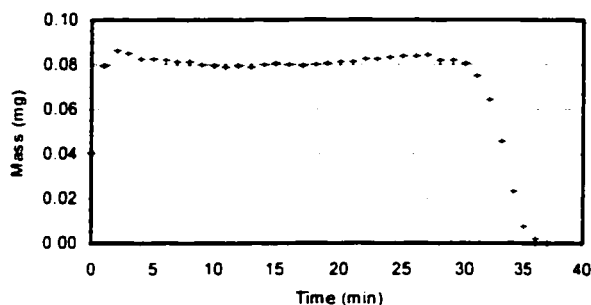
Case 1, run 1 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	27	1	1.515	-0.020	1.515	0.071	0.006	0.047	0.004	-	-
	2	27	1	1.515	-0.020	1.515	0.071	0.006	0.047	0.004	0.047	0.004
	3	27	1	1.515	0.020	1.515	0.071	0.006	0.047	0.004	0.047	0.004
	4	-	26	1.517	-0.020	1.517	0.071	0.004	0.047	0.003	0.047	0.003

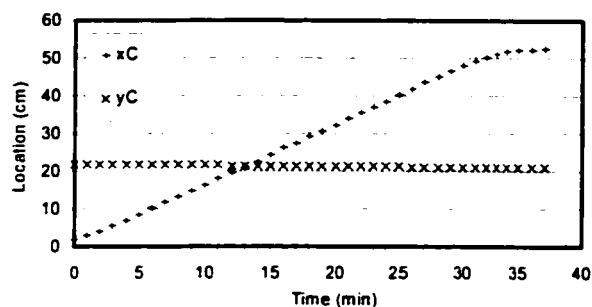
Homogeneous Medium - Laboratory Results

Case 1, run 2 (Steady State, $i = 0.1211$)

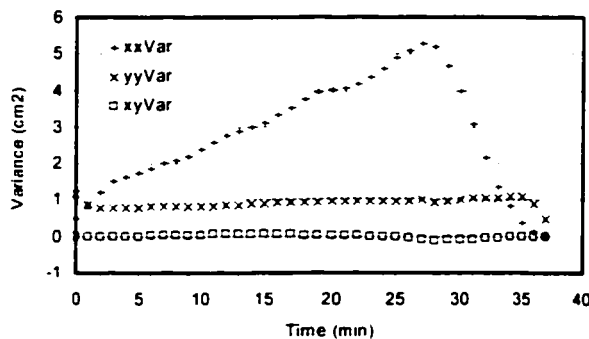
Case 1 (Steady State) - Run 2
Mass vs. Time



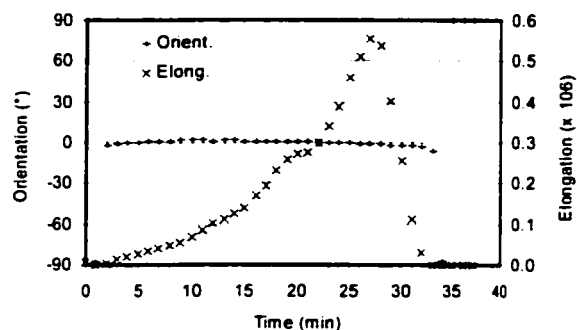
Case 1 (Steady State) - Run 2
Centroid Location vs. Time



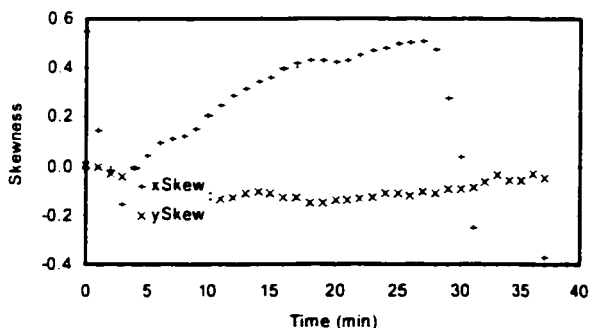
Case 1 (Steady State) - Run 2
Variance vs. Time



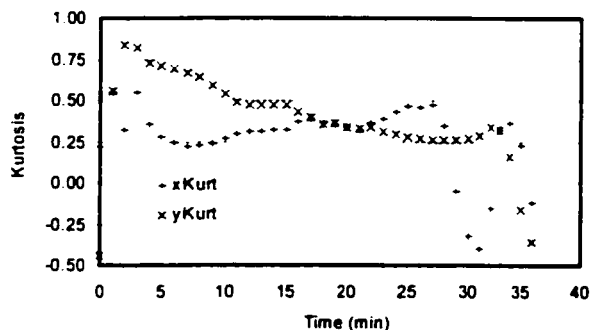
Case 1 (Steady State) - Run 2
Orientation & Elongation vs. Time



Case 1 (Steady State) - Run 2
Skewness vs. Time



Case 1 (Steady State) - Run 2
Kurtosis vs. Time

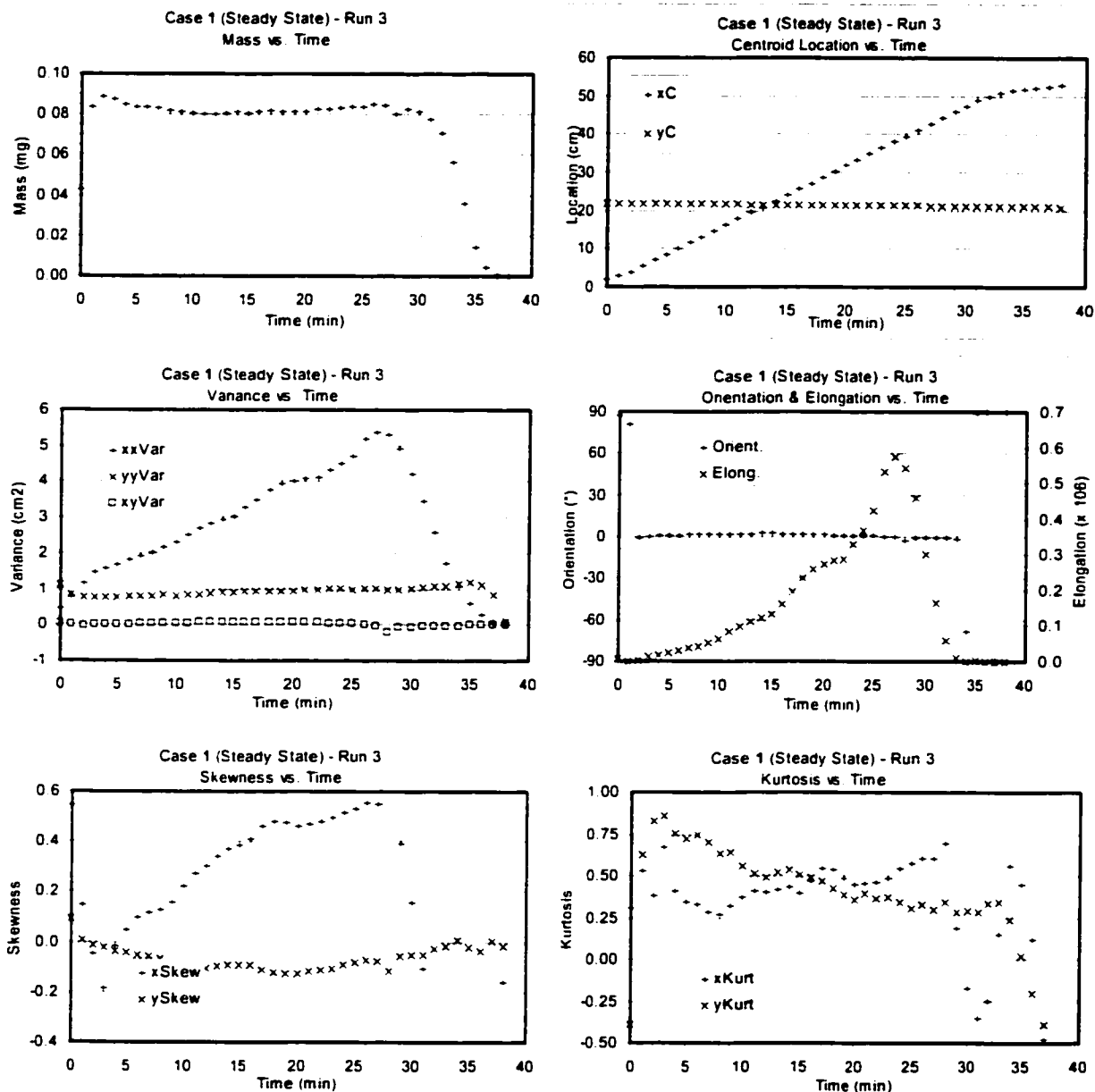


Case 1, run 2 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	25	1	1.583	-0.023	1.583	0.078	0.005	0.049	0.003	-	-
	2	25	1	1.583	-0.023	1.583	0.078	0.005	0.049	0.003	0.049	0.003
	3	25	1	1.583	0.023	1.583	0.078	0.005	0.049	0.003	0.049	0.003
	4	-	24	1.585	-0.024	1.585	0.079	0.005	0.050	0.003	0.050	0.003

Homogeneous Medium – Laboratory Results

Case 1, run 3 (Steady State, $i = 0.1211$)

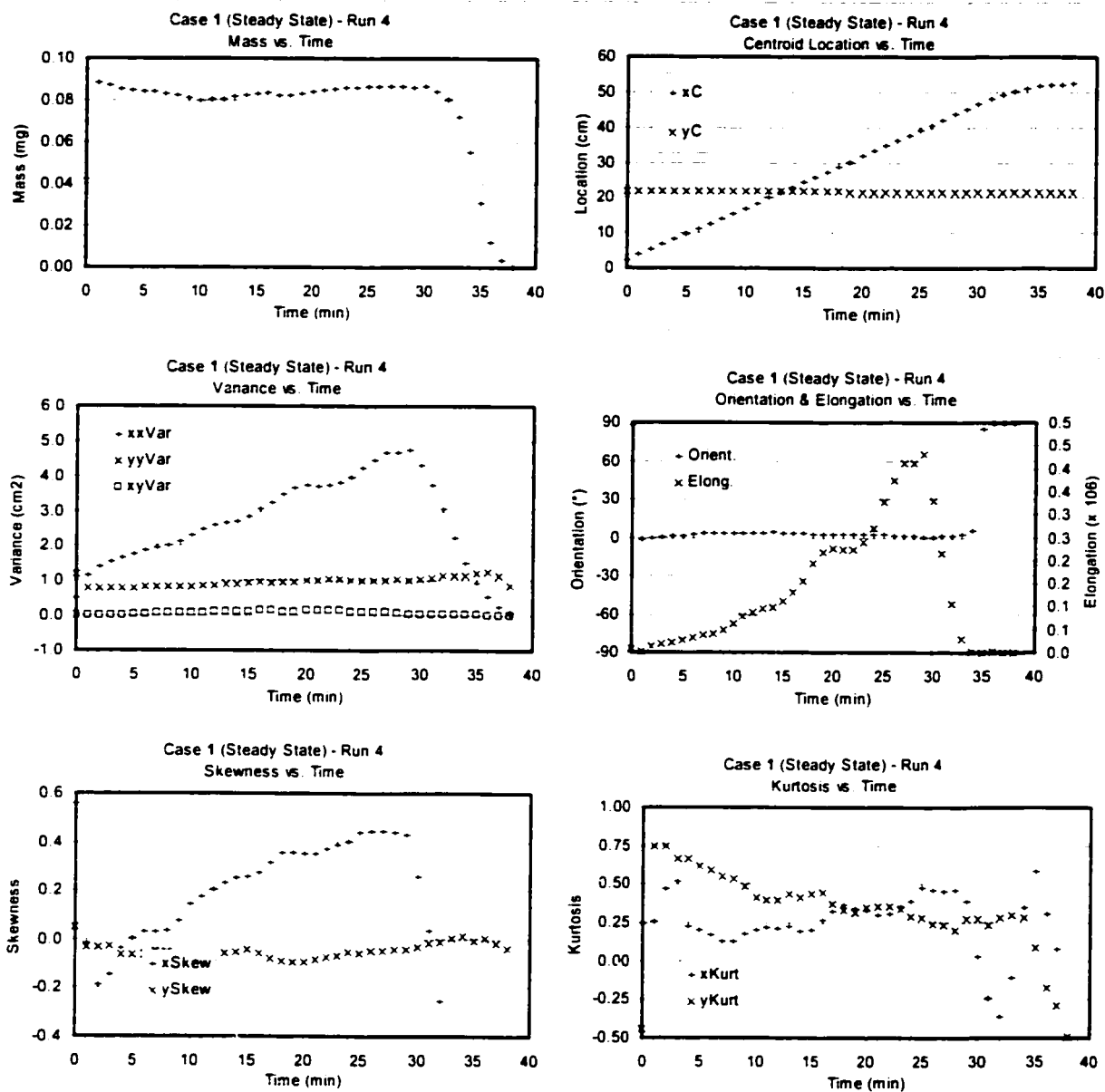


Case 1, run 3 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	25	1	1.554	-0.018	1.554	0.079	0.006	0.051	0.004	-	-
	2	25	1	1.554	-0.018	1.554	0.079	0.006	0.051	0.004	0.051	0.004
	3	25	1	1.554	0.018	1.554	0.079	0.006	0.051	0.004	0.051	0.004
	4	-	24	1.556	-0.018	1.556	0.081	0.005	0.052	0.003	0.052	0.003

Homogeneous Medium - Laboratory Results

Case 1, run 4 (Steady State, $i = 0.1211$)

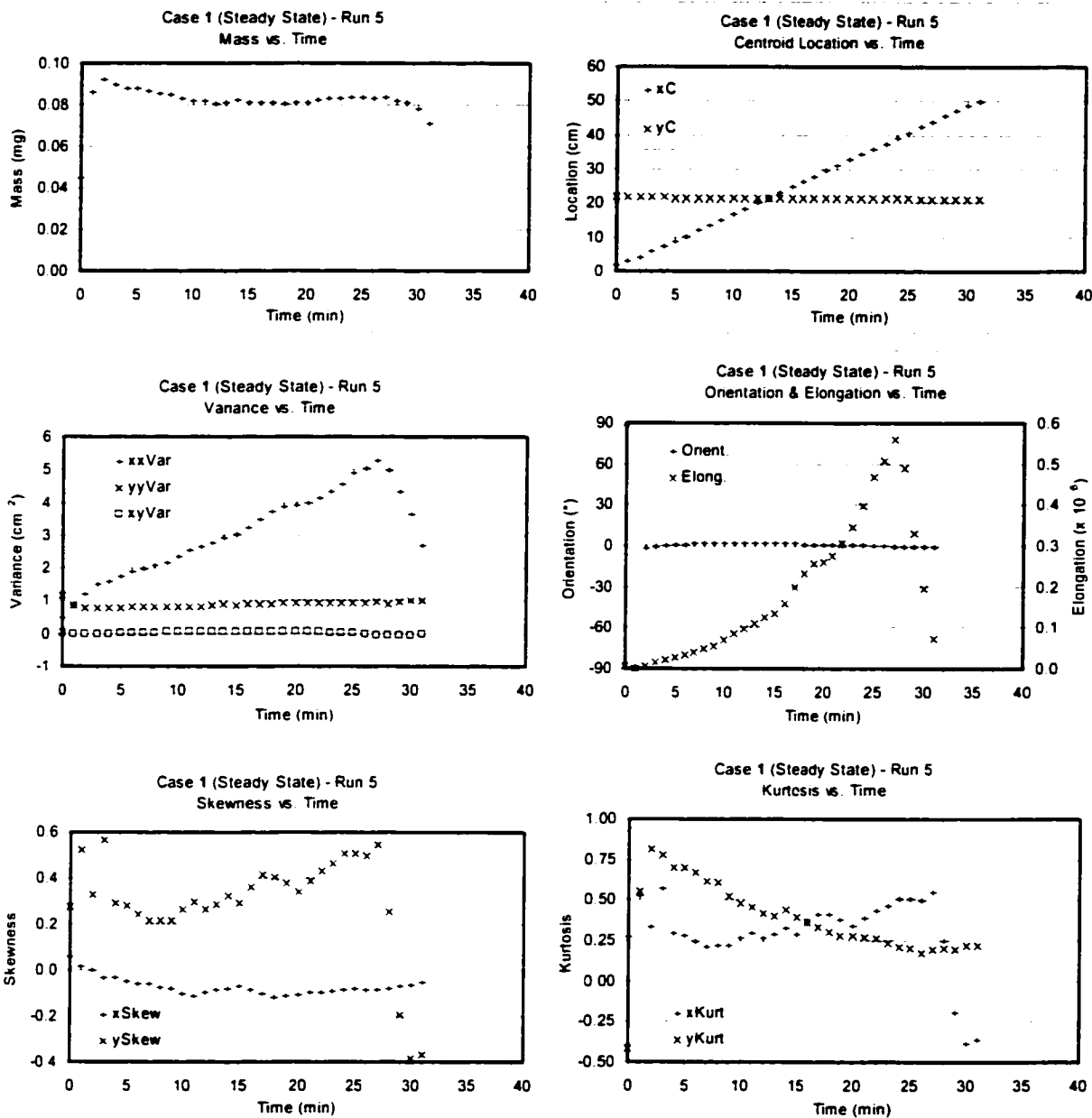


Case 1, run 4 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$ v $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	27	1	1.481	-0.010	1.481	0.063	0.005	0.043	0.004	-	-
	2	27	1	1.481	-0.010	1.481	0.063	0.005	0.043	0.004	0.043	0.004
	3	27	1	1.481	0.010	1.481	0.063	0.005	0.043	0.004	0.043	0.004
	4	-	26	1.483	-0.009	1.483	0.061	0.004	0.041	0.003	0.041	0.003

Homogeneous Medium – Laboratory Results

Case 1, run 5 (Steady State, $i = 0.1211$)

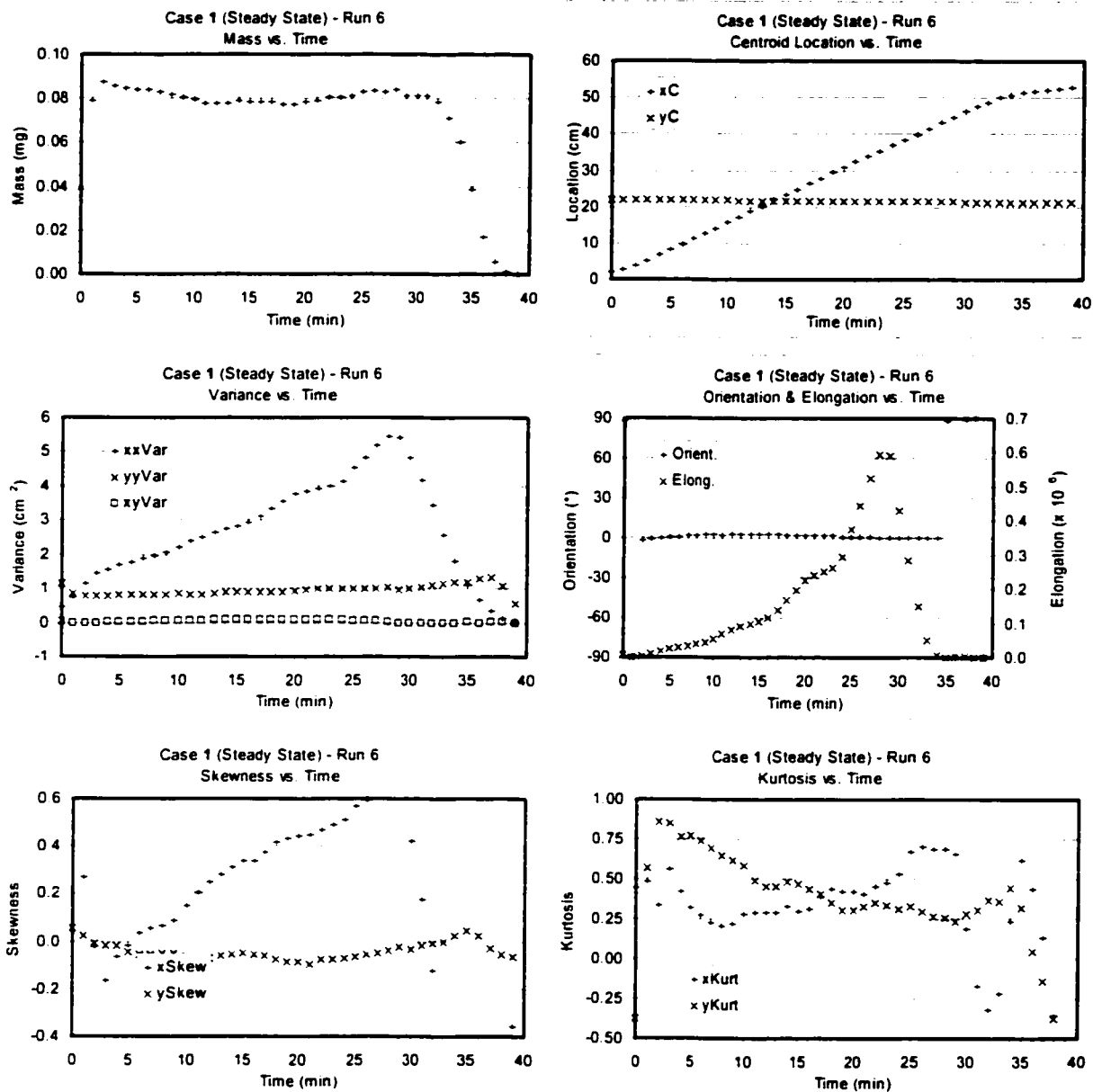


Case 1, run 5 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$ v $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	25	1	1.598	-0.019	1.599	0.077	0.004	0.048	0.002	-
	2	25	1	1.598	-0.019	1.599	0.077	0.004	0.048	0.002	0.048
	3	25	1	1.598	0.019	1.599	0.077	0.004	0.048	0.002	0.048
	4	-	24	1.599	-0.018	1.599	0.071	0.003	0.044	0.002	0.044

Homogeneous Medium – Laboratory Results

Case 1, run 6 (Steady State, $i = 0.1211$)

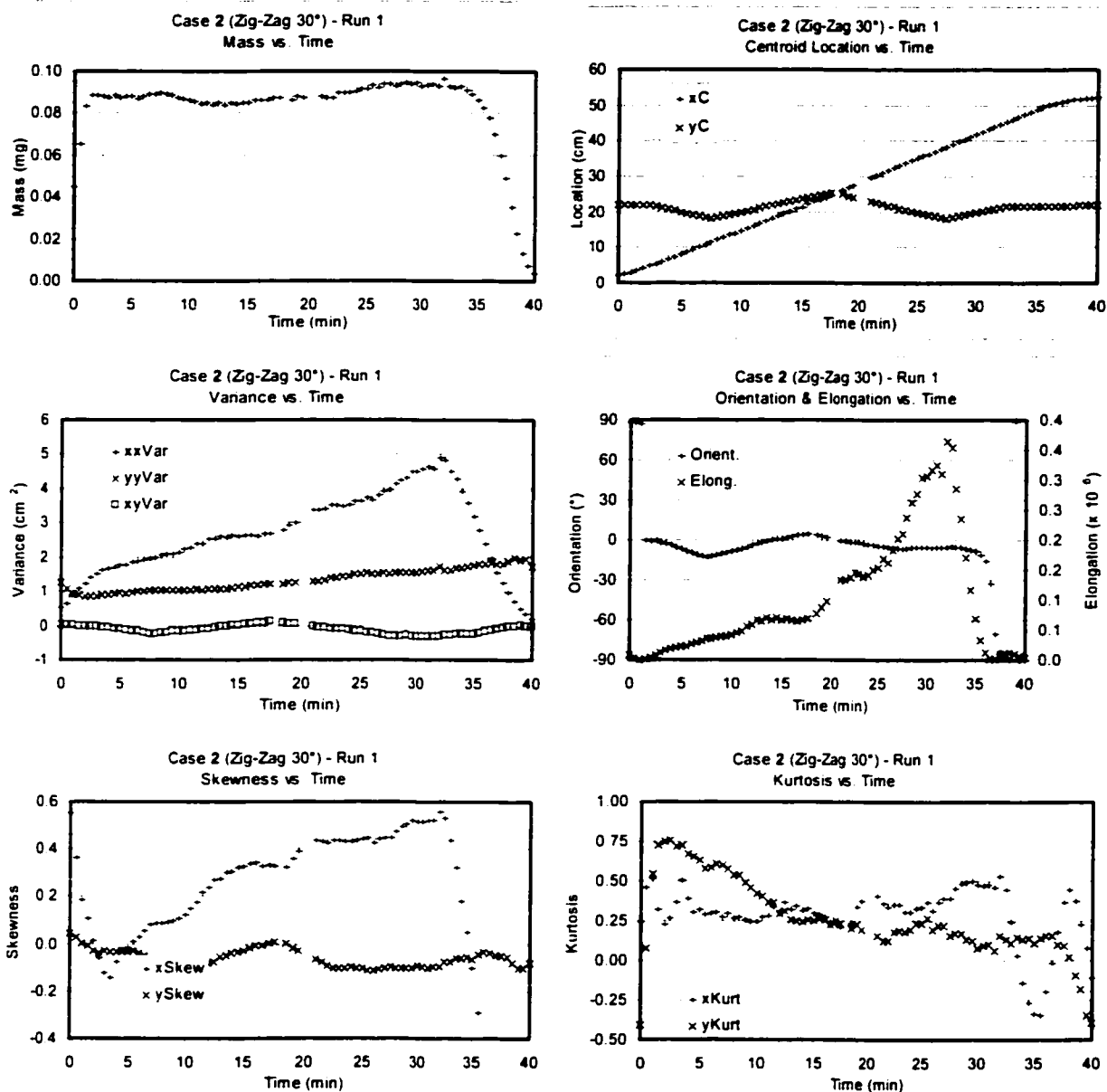


Case 1, run 6 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min^{-1}	cm min^{-1}	cm min^{-1}	$\text{cm}^2 \text{ min}^{-1}$	$\text{cm}^2 \text{ min}^{-1}$	cm	cm	cm	cm
Method	1	26	1	1.512	-0.014	1.512	0.075	0.005	0.050	0.003	-
	2	26	1	1.512	-0.014	1.512	0.075	0.005	0.050	0.003	0.050
	3	26	1	1.512	0.014	1.512	0.075	0.005	0.050	0.003	0.050
	4	-	25	1.513	-0.013	1.513	0.081	0.005	0.053	0.003	0.053

Homogeneous Medium – Laboratory Results

Case 2, run 1 (Zigzag 30°, $i = 0.1211$)

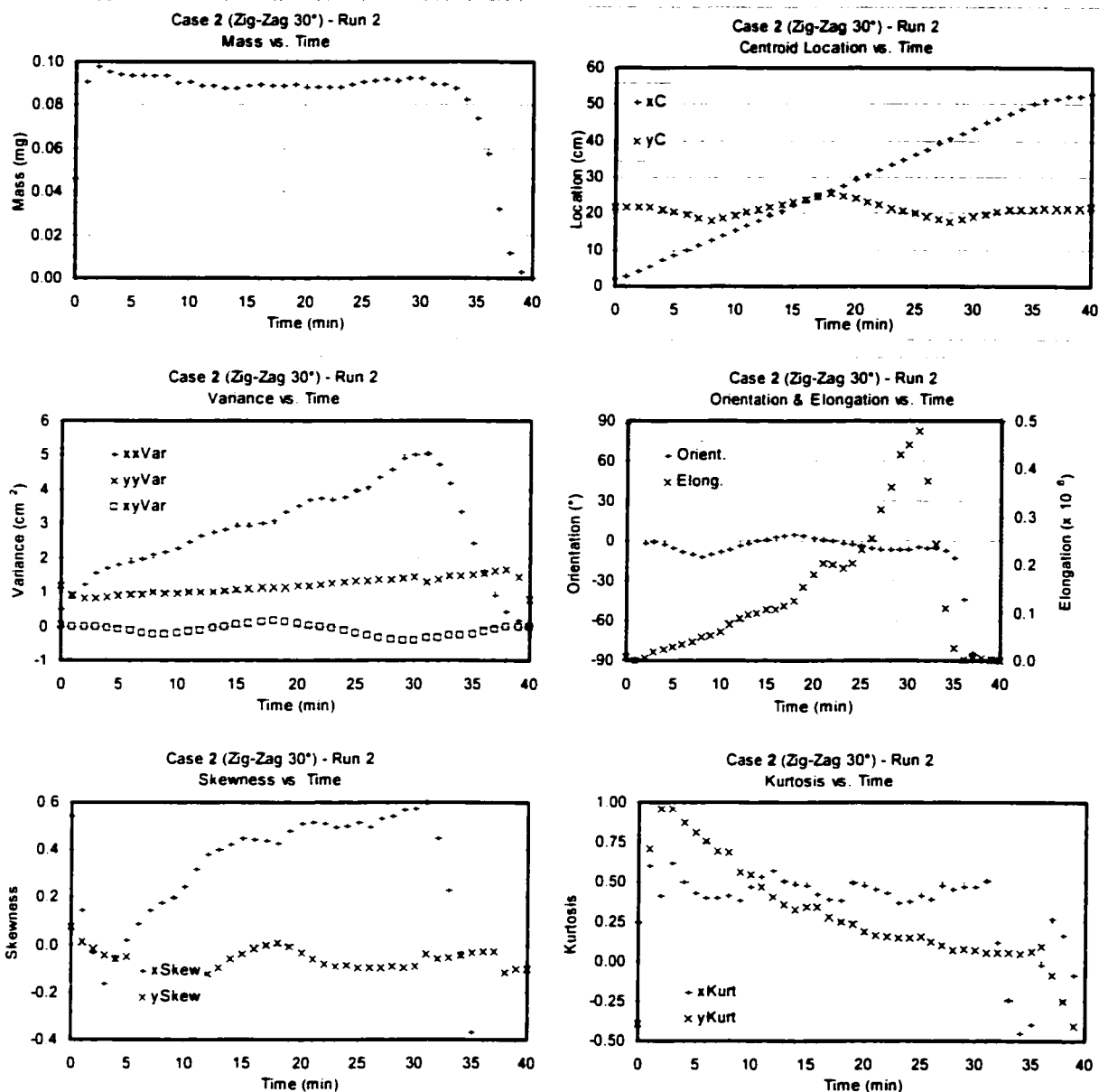


Case 2, run 1 (Homogeneous medium, Zig-Zag 30° flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$ v $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	55	1.354	-0.013	1.354	0.053	0.013	0.039	0.010	-	-
	2	13	1.360	-0.021	1.361	0.053	0.015	0.039	0.011	0.034	0.009
	3	13	1.360	0.754	1.555	0.053	0.015	0.034	0.009	0.034	0.009
	4	-	54	1.357	-0.022	1.358	0.054	0.013	0.040	0.010	0.035

Homogeneous Medium - Laboratory Results

Case 2, run 2 (Zigzag 30°, $i = 0.1211$)

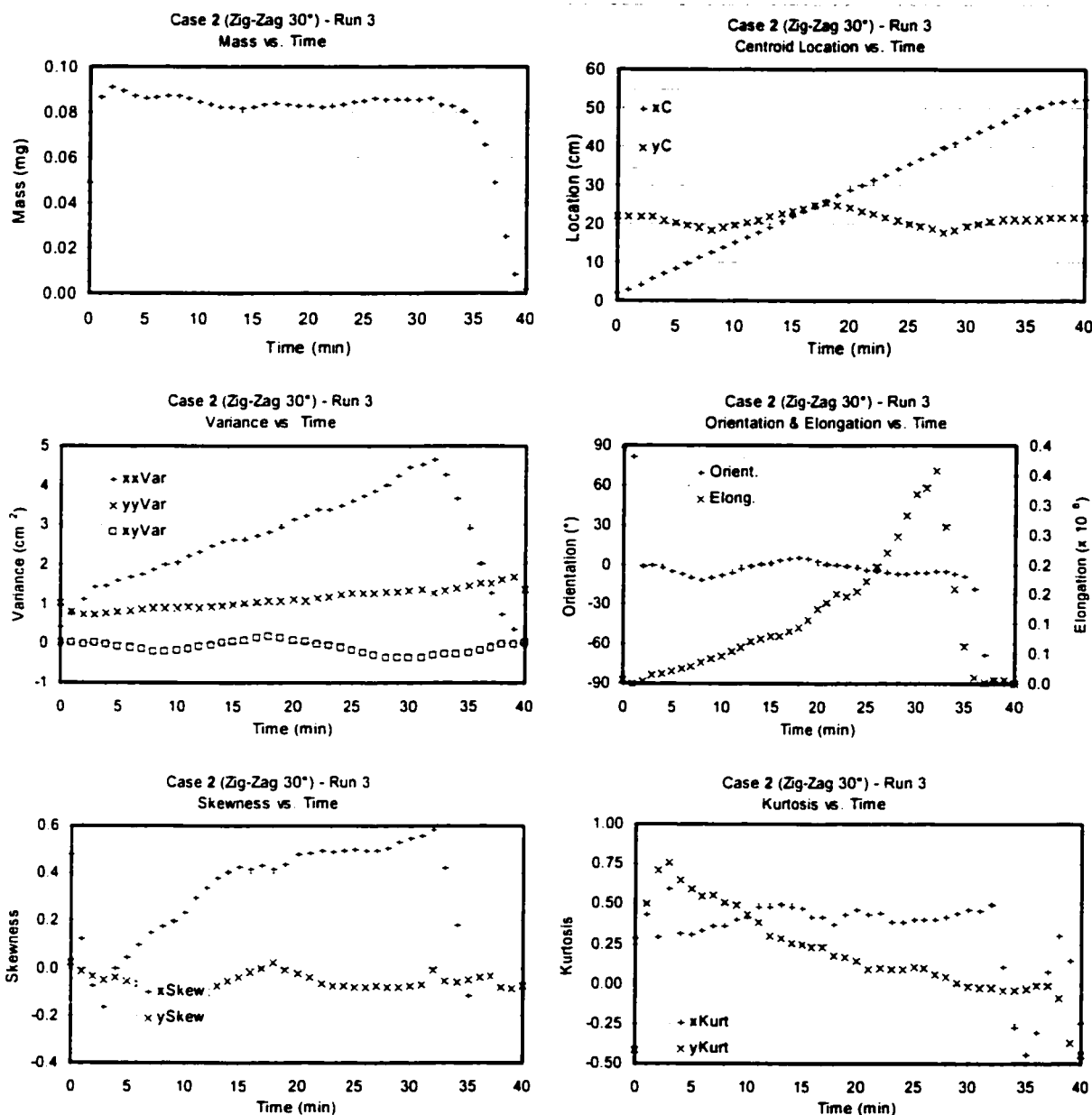


Case 2, run 2 (Homogeneous medium, Zig-Zag 30° flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	28	1	1.396	-0.022	1.396	0.060	0.010	0.043	0.007	-	-
	2	7	4	1.403	-0.074	1.405	0.058	0.009	0.041	0.007	0.036	0.006
	3	7	4	1.403	0.754	1.593	0.058	0.009	0.037	0.006	0.036	0.006
	4	-	27	1.402	-0.071	1.403	0.063	0.008	0.045	0.006	0.039	0.005

Homogeneous Medium – Laboratory Results

Case 2, run 3 (Zigzag 30°, $i = 0.1211$)

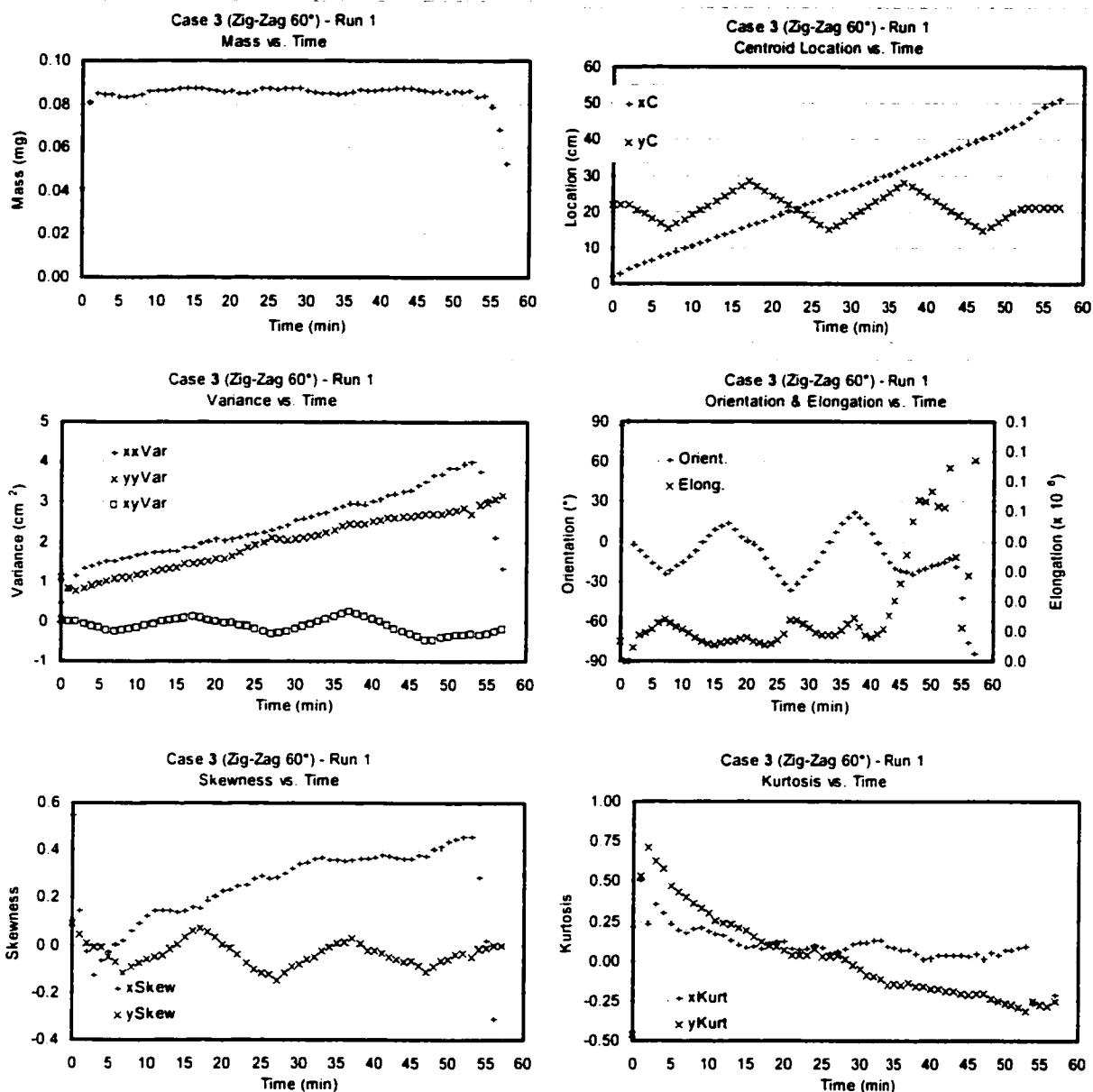


Case 2, run 3 (Homogeneous medium, Zig-Zag 30° flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	29	1	1.357	-0.020	1.357	0.055	0.011	0.040	0.008	-	-
	2	7	4	1.363	-0.040	1.364	0.055	0.010	0.040	0.008	0.035	0.007
	3	7	4	1.363	0.736	1.549	0.055	0.010	0.035	0.007	0.035	0.007
	4	-	28	1.362	-0.039	1.363	0.056	0.009	0.041	0.007	0.036	0.006

Homogeneous Medium – Laboratory Results

Case 3, run 1 (Zigzag 60°, $i = 0.1211$)

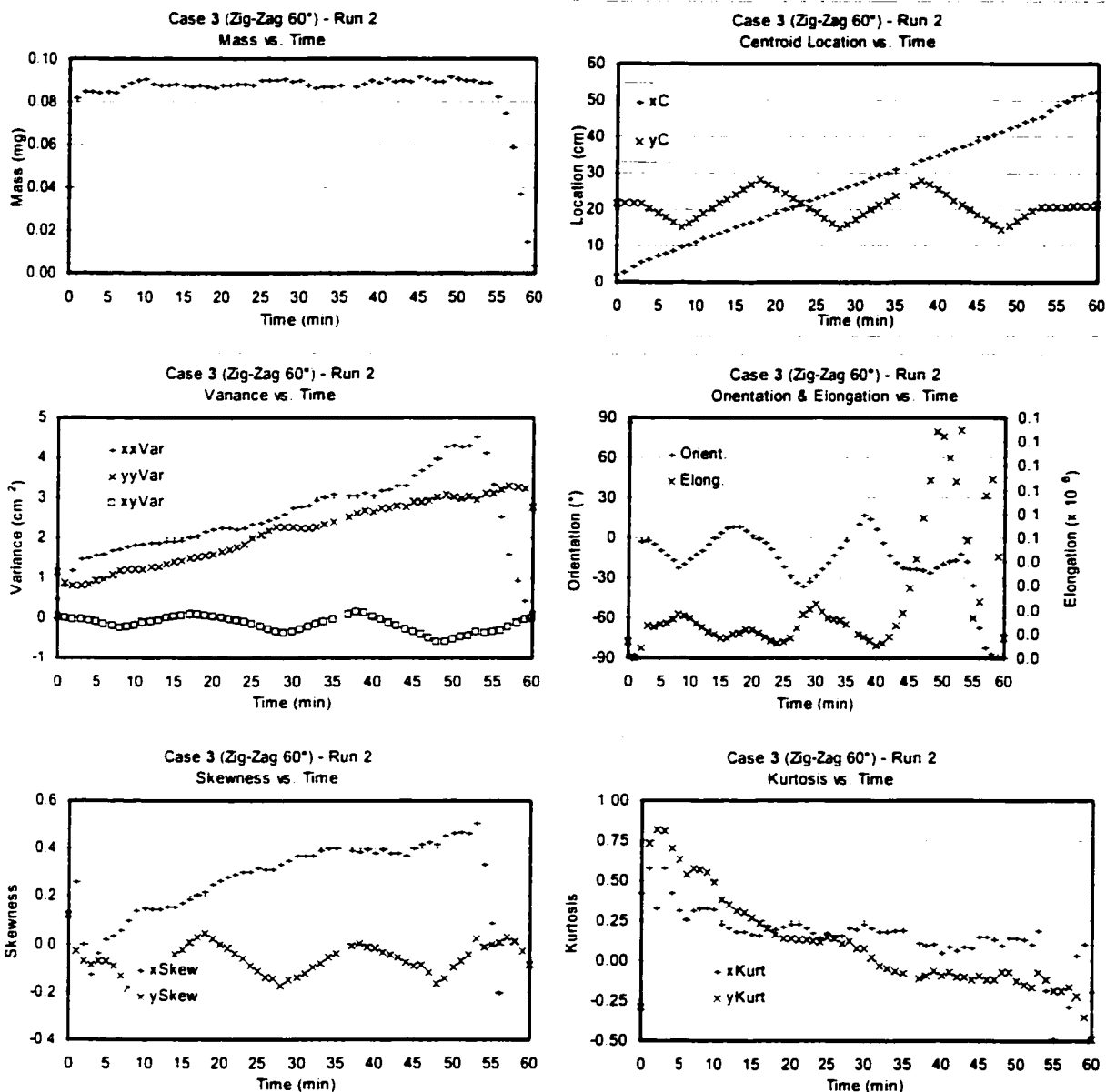


Case 3, run 1 (Homogeneous medium, Zig-Zag 60° flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	49	1	0.803	-0.017	0.803	0.025	0.021	0.032	0.026	-	-
	2	8	6	0.807	0.035	0.808	0.026	0.021	0.032	0.026	0.017	0.014
	3	8	6	0.807	1.310	1.539	0.026	0.021	0.017	0.014	0.017	0.014
	4	-	48	0.806	0.034	0.807	0.026	0.020	0.033	0.025	0.017	0.013

Homogeneous Medium – Laboratory Results

Case 3, run 2 (Zigzag 60°, $i = 0.1211$)

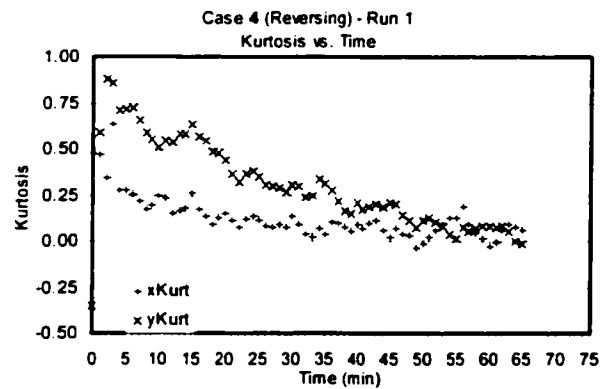
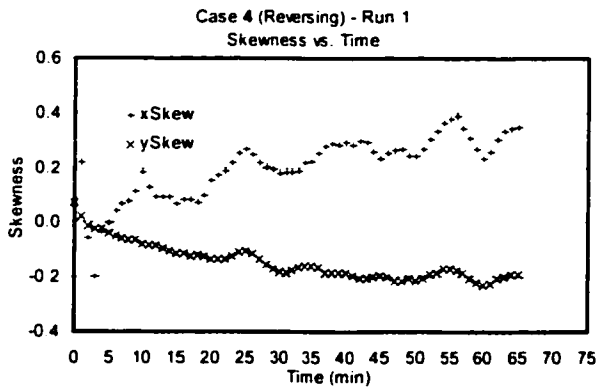
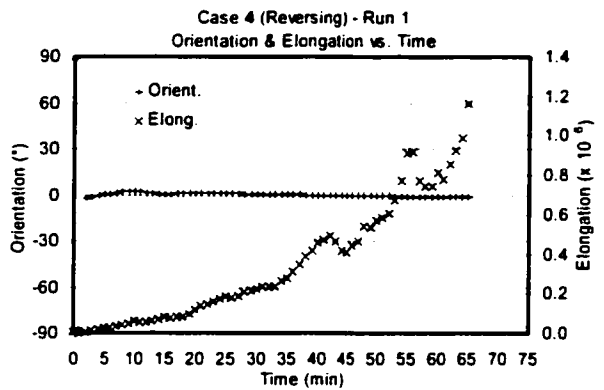
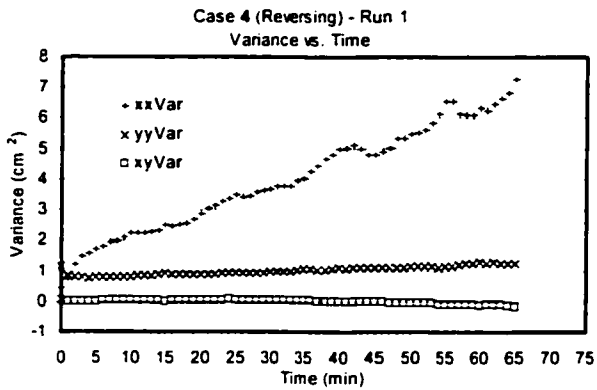
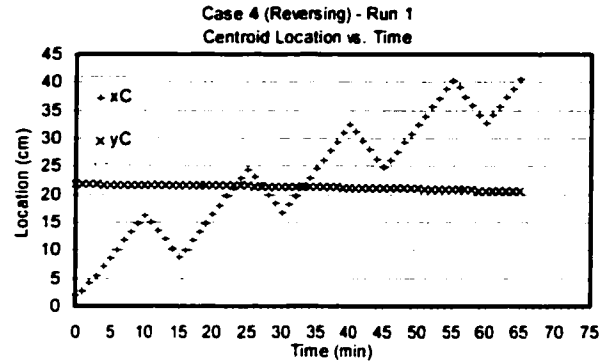
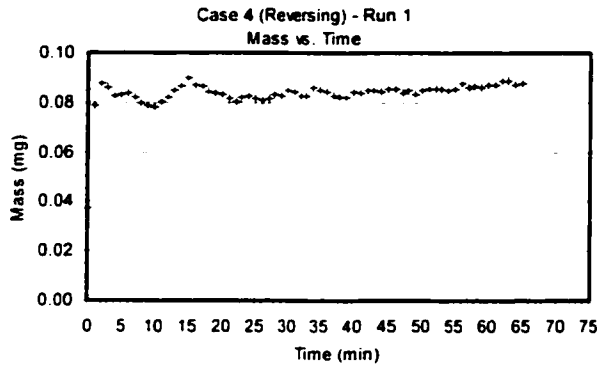


Case 3, run 2 (Homogeneous medium, Zig-Zag 60° flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	48	1	0.797	-0.021	0.797	0.028	0.024	0.035	0.030	-	-
	2	8	6	0.801	-0.019	0.801	0.028	0.023	0.034	0.028	0.018	0.015
	3	8	6	0.801	1.315	1.540	0.028	0.023	0.018	0.015	0.018	0.015
	4	-	47	0.802	-0.021	0.802	0.029	0.023	0.036	0.029	0.019	0.015

Homogeneous Medium – Laboratory Results

Case 4, run 1 (Reversing, $i = 0.1211$)

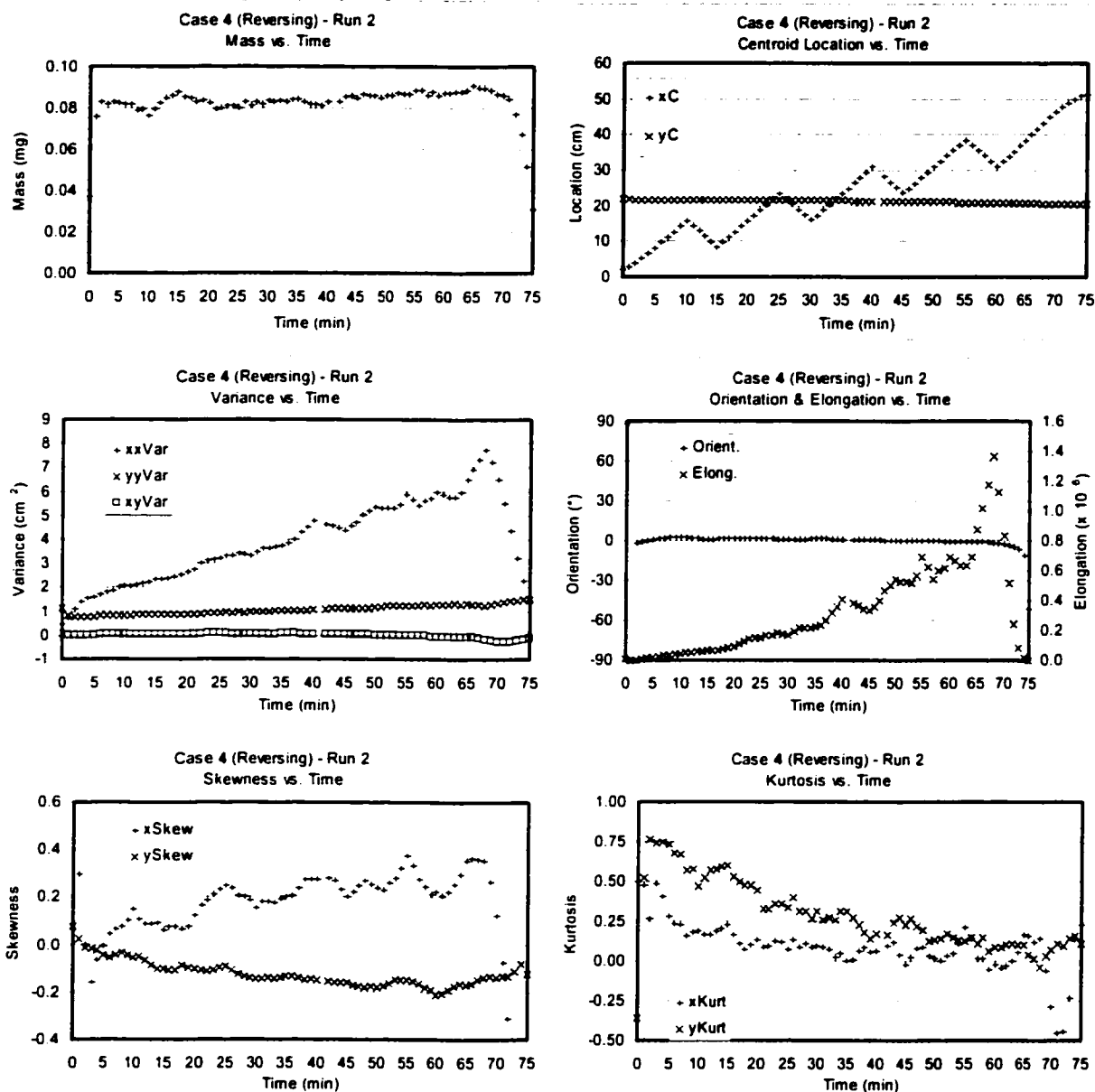


Case 4, run 1 (Homogeneous medium, Reversing flow pattern, $i = 0.1211$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	61	1	0.508	-0.019	0.508	0.044	0.004	0.086	0.008	-	-
	2	6	9	0.554	-0.019	0.554	0.045	0.004	0.081	0.007	0.029	0.002
	3	6	9	1.565	0.019	1.565	0.045	0.004	0.029	0.002	0.029	0.002
	4	-	60	0.550	-0.019	0.550	0.047	0.004	0.085	0.007	0.028	0.002

Homogeneous Medium - Laboratory Results

Case 4, run 2 (Reversing, $i = 0.1211$)

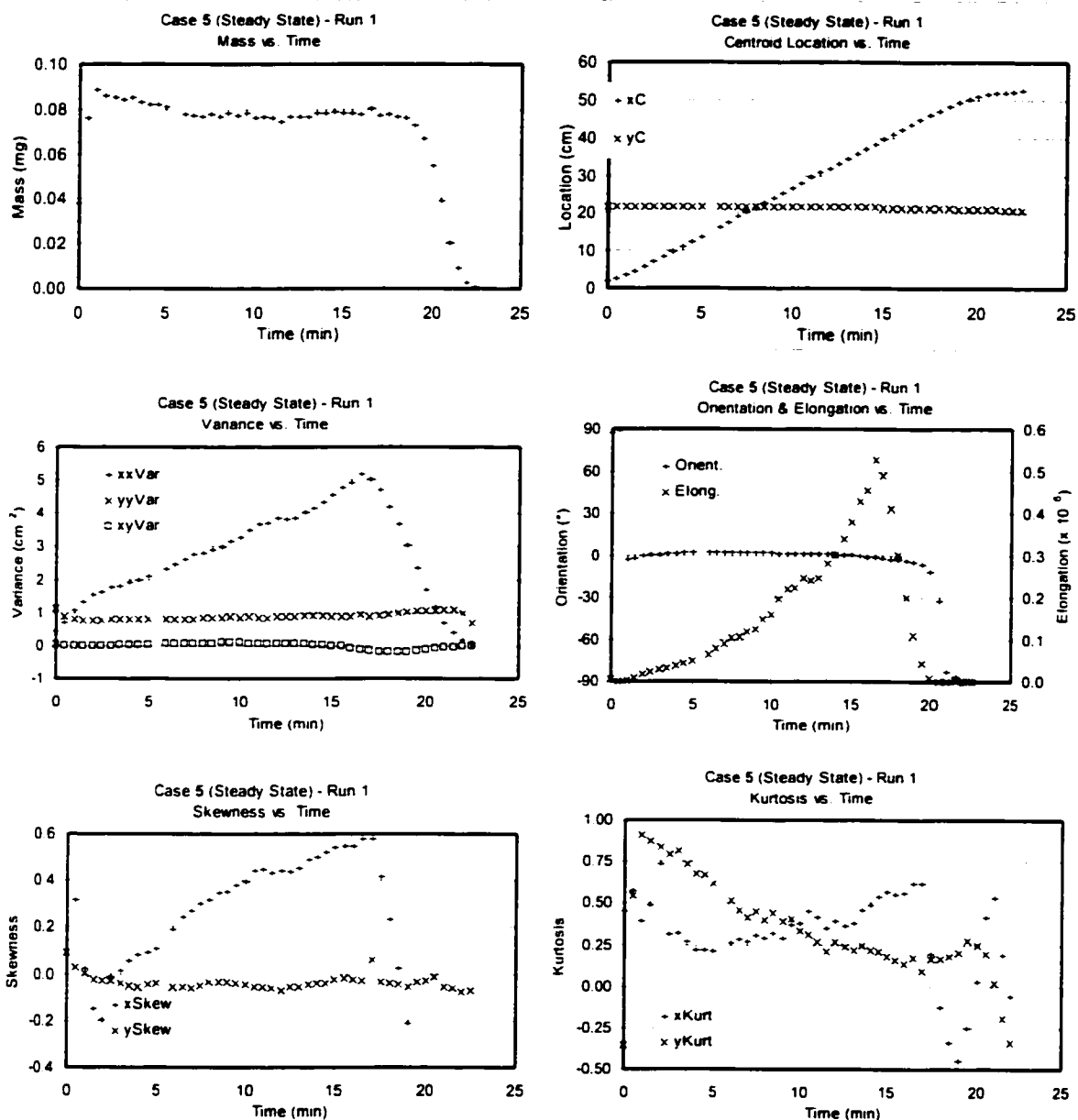


Case 4, run 2 (Homogeneous medium, Reversing flow pattern, $i = 0.1211$)

Table 4. Analysis of the motion of the electron beam, reversing flow pattern. (Contd.)												
		N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
				cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	63	1	0.493	-0.016	0.493	0.041	0.004	0.082	0.009	-	-
	2	7	9	0.576	-0.016	0.576	0.048	0.004	0.083	0.006	0.032	0.002
	3	7	9	1.507	0.016	1.507	0.048	0.004	0.032	0.002	0.032	0.002
	4	-	62	0.572	-0.016	0.572	0.049	0.004	0.085	0.006	0.031	0.002

Homogeneous Medium – Laboratory Results

Case 5, run 1 (Steady State, $i = 0.2174$)

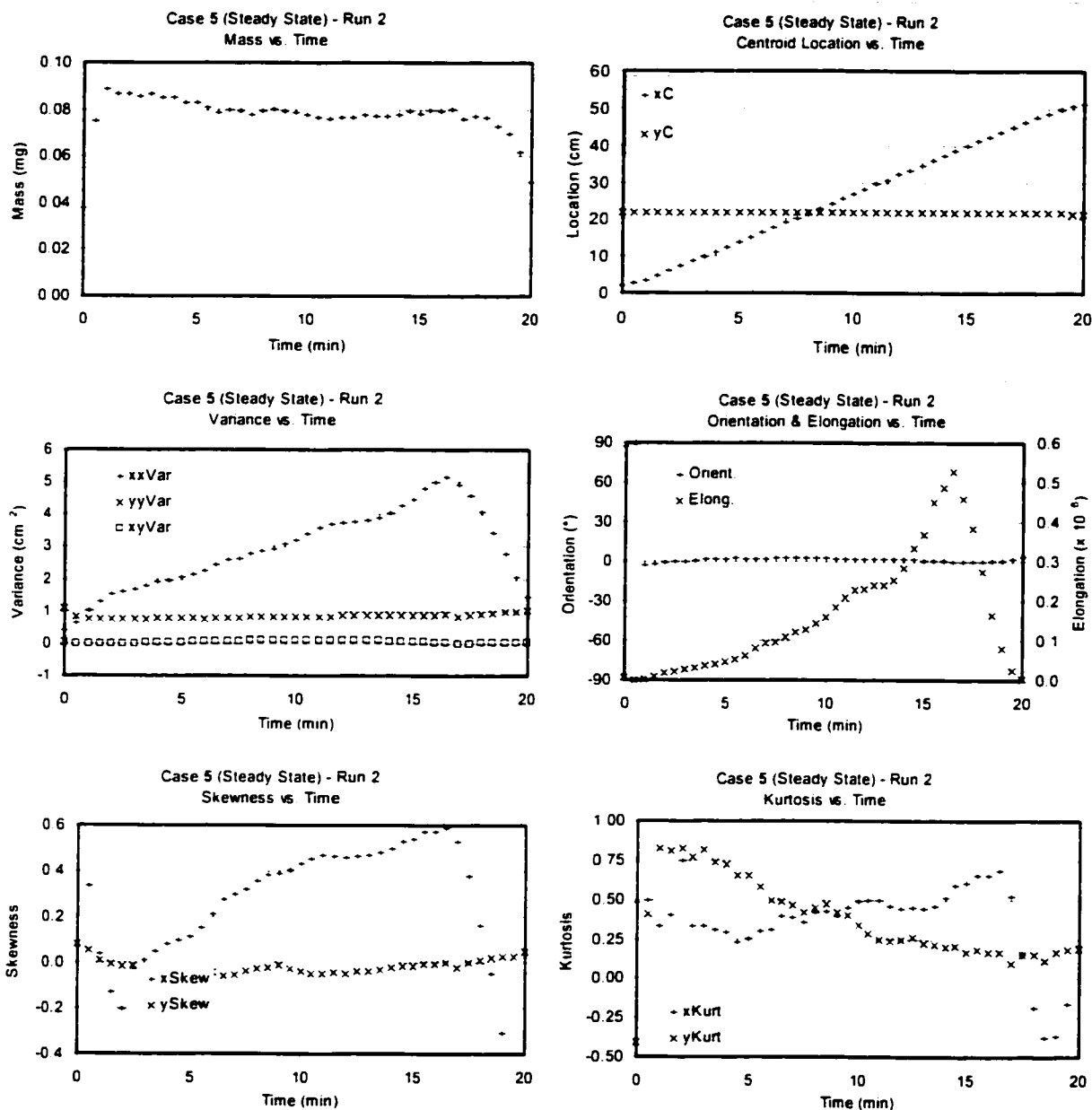


Case 5, run 1 (Steady State, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	29	1	2.590	-0.018	2.590	0.120	0.005	0.046	0.002	-	-
	2	29	1	2.590	-0.018	2.590	0.120	0.005	0.046	0.002	0.046	0.002
	3	29	1	2.590	0.018	2.590	0.120	0.005	0.046	0.002	0.046	0.002
	4	-	28	2.588	-0.020	2.588	0.126	0.006	0.049	0.002	0.049	0.002

Homogeneous Medium – Laboratory Results

Case 5, run 2 (Steady State, $i = 0.2174$)

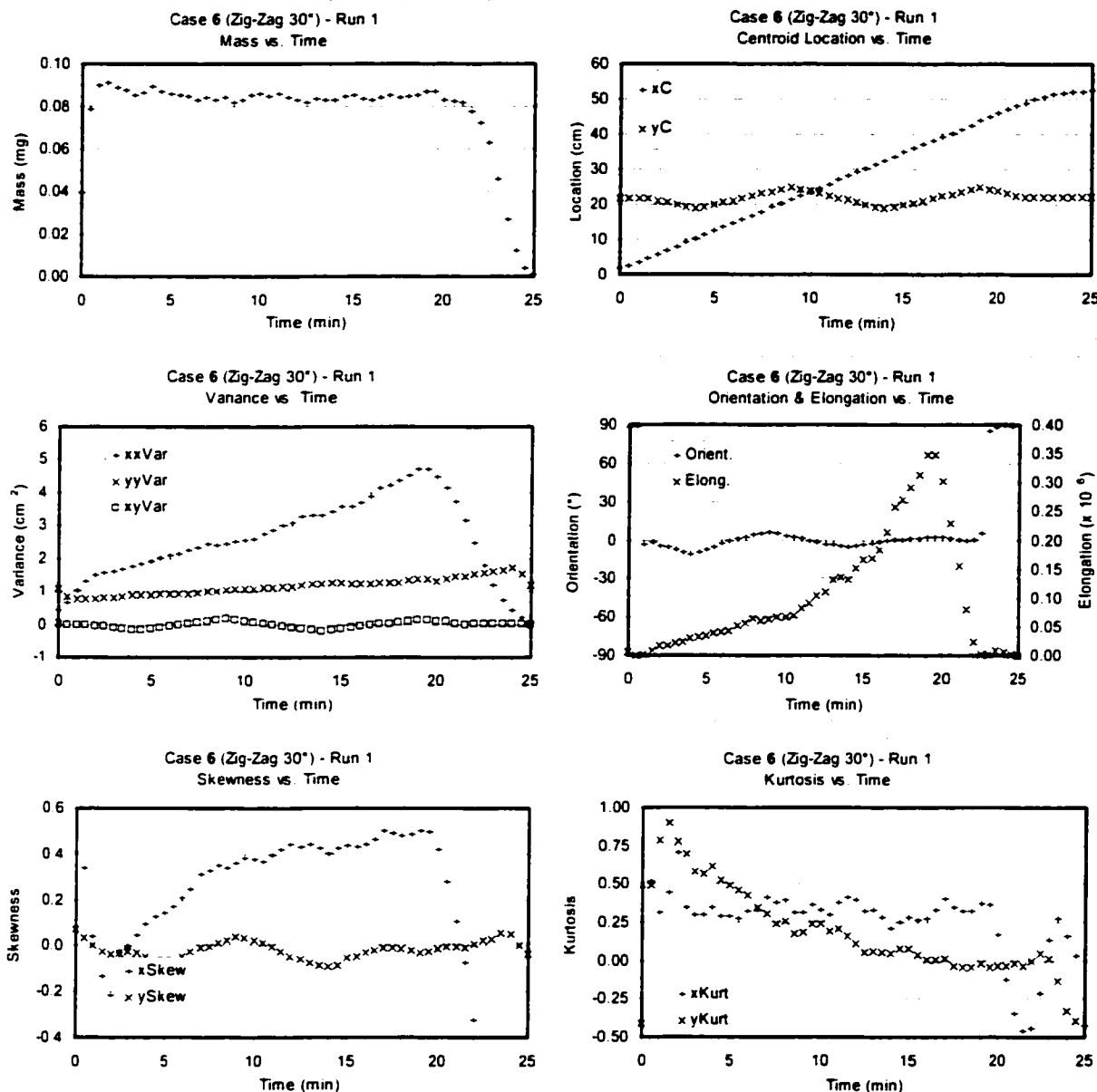


Case 5, run 2 (Steady State, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min^{-1}	cm min^{-1}	cm min^{-1}	$\text{cm}^2 \text{min}^{-1}$	$\text{cm}^2 \text{min}^{-1}$	cm	cm	cm	cm
Method	1	30	1	2.601	-0.009	2.601	0.120	0.005	0.046	0.002	-
	2	30	1	2.601	-0.009	2.601	0.120	0.005	0.046	0.002	0.046
	3	30	1	2.601	0.009	2.601	0.120	0.005	0.046	0.002	0.046
	4	-	29	2.608	-0.008	2.608	0.126	0.006	0.048	0.002	0.048

Homogeneous Medium – Laboratory Results

Case 6, run 1 (Zigzag 30°, $i = 0.2174$)

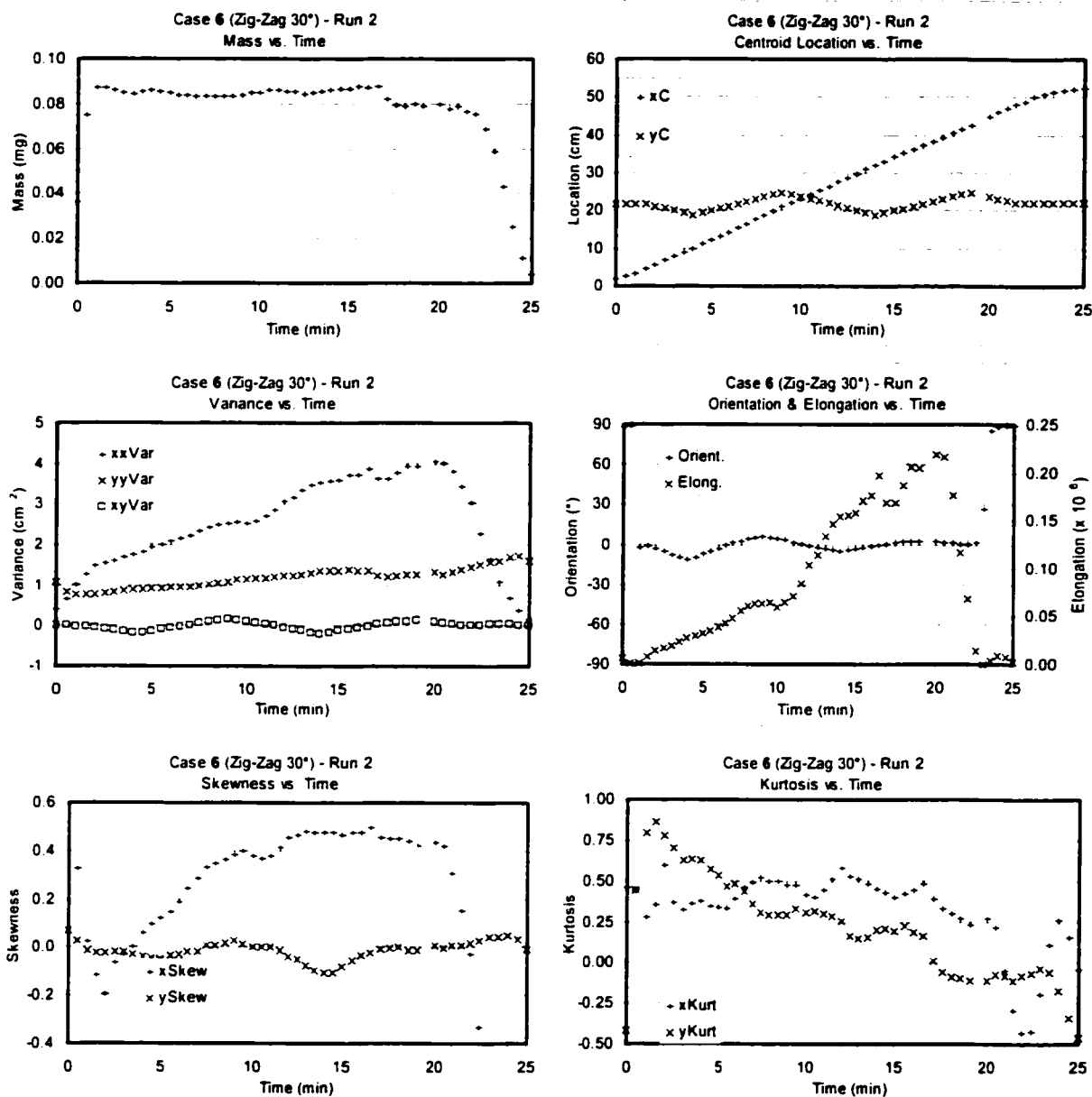


Case 6, run 1 (Zig-Zag 30°, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	35	1	2.238	0.100	2.240	0.088	0.016	0.039	0.007	-
	2	8	4	2.238	0.182	2.245	0.101	0.015	0.045	0.007	0.040
	3	8	4	2.238	1.223	2.550	0.101	0.015	0.040	0.006	0.040
	4	-	34	2.236	0.213	2.246	0.094	0.018	0.042	0.008	0.037

Homogeneous Medium - Laboratory Results

Case 6, run 2 (Zigzag 30°, $i = 0.2174$)

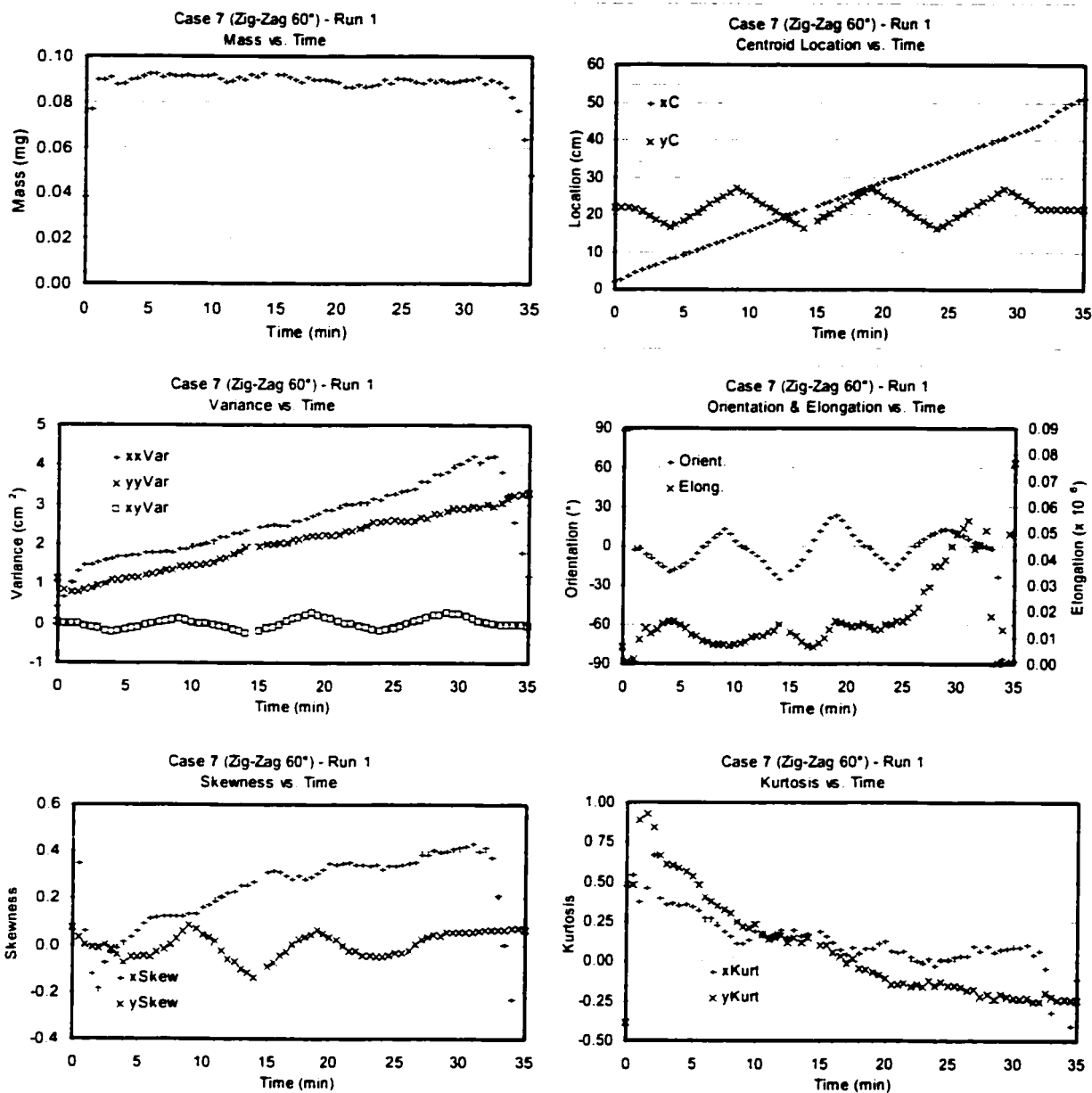


Case 6, run 2 (Zig-Zag 30°, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	36	1	2.183	0.113	2.186	0.076	0.016	0.035	0.007	-
	2	7	5	2.175	0.115	2.178	0.078	0.012	0.036	0.006	0.031
	3	7	5	2.175	1.192	2.481	0.078	0.012	0.031	0.005	0.031
	4	-	35	2.180	0.143	2.184	0.072	0.015	0.033	0.007	0.029

Homogeneous Medium – Laboratory Results

Case 7, run 1 (Zigzag 60°, $i = 0.2174$)

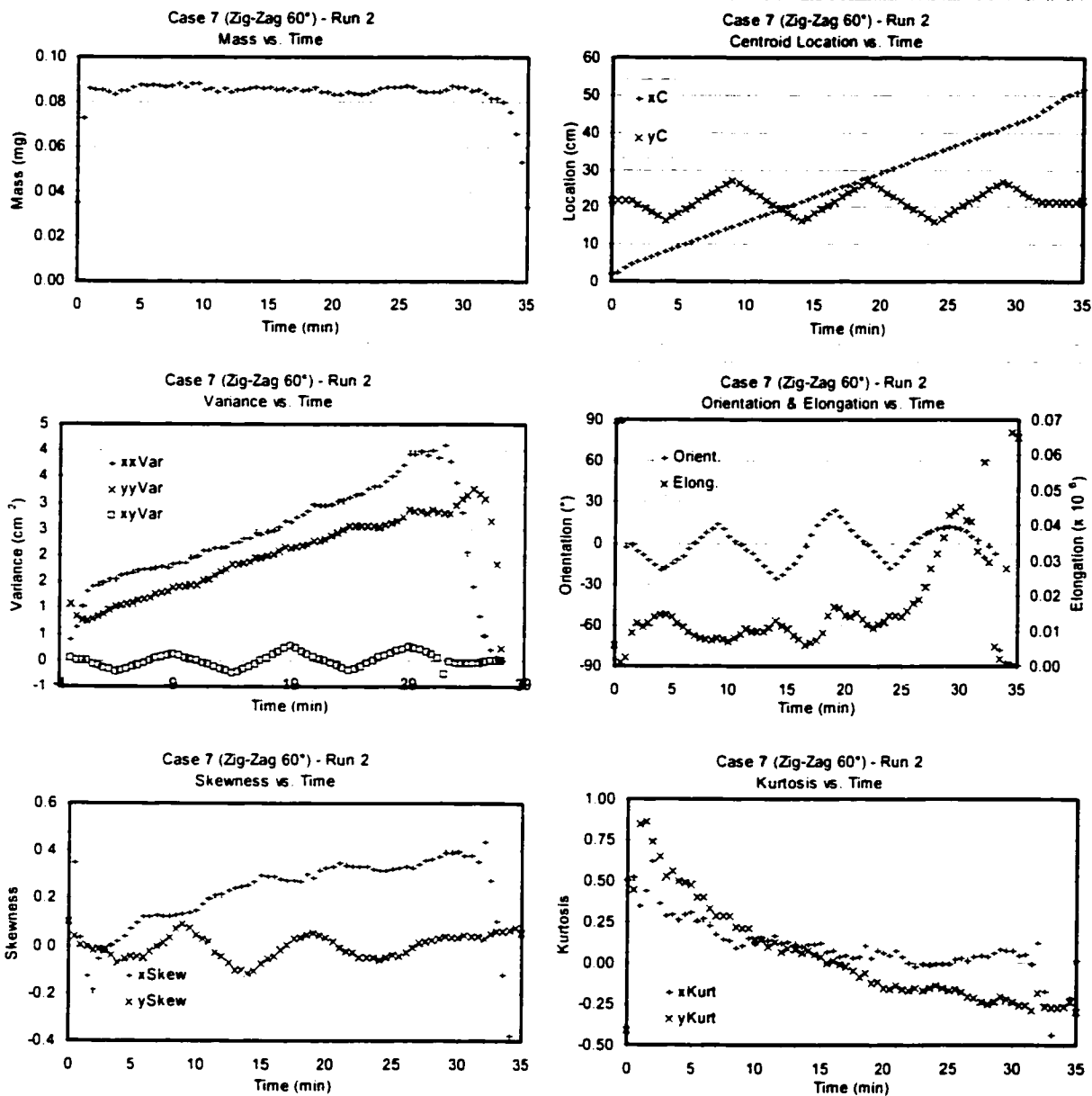


Case 7, run 1 (Zig-Zag 60°, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	59	1.306	0.078	1.308	0.045	0.036	0.034	0.027	–	–
	2	8	1.313	-0.006	1.313	0.045	0.035	0.034	0.027	0.018	0.014
	3	8	1.313	2.112	2.487	0.045	0.035	0.018	0.014	0.018	0.014
	4	–	58	1.309	0.032	1.309	0.044	0.035	0.034	0.027	0.018

Homogeneous Medium - Laboratory Results

Case 7, run 2 (Zigzag 60°, $i = 0.2174$)

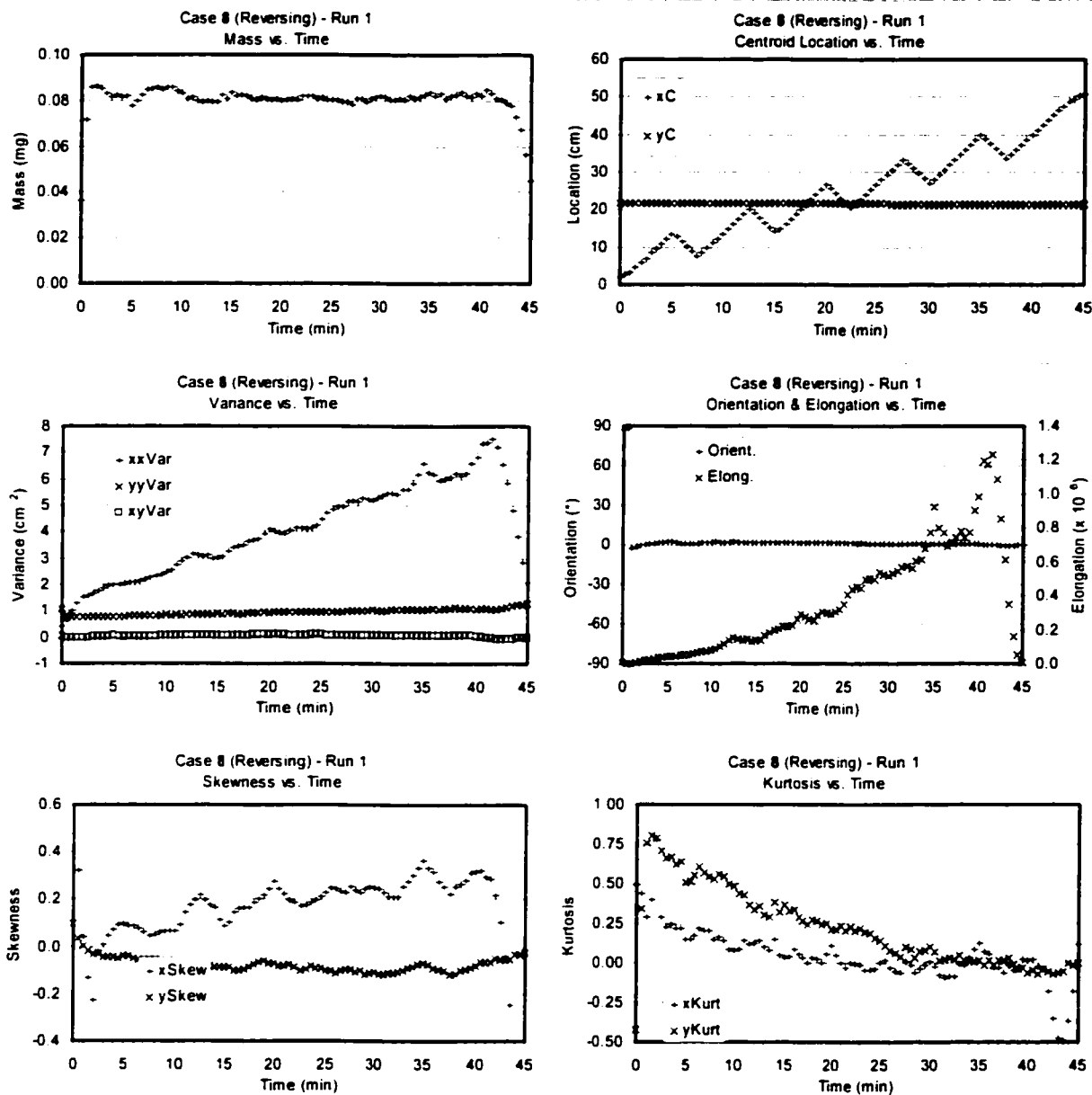


Case 7, run 2 (Zig-Zag 60°, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	59	1	1.322	0.081	1.324	0.044	0.036	0.033	0.027	-
	2	8	7	1.330	0.069	1.332	0.042	0.035	0.032	0.026	0.017
	3	8	7	1.330	2.166	2.542	0.042	0.035	0.017	0.014	0.017
	4	-	58	1.325	0.069	1.327	0.044	0.036	0.034	0.027	0.018

Homogeneous Medium - Laboratory Results

Case 8, run 1 (Reversing, $i = 0.2174$)

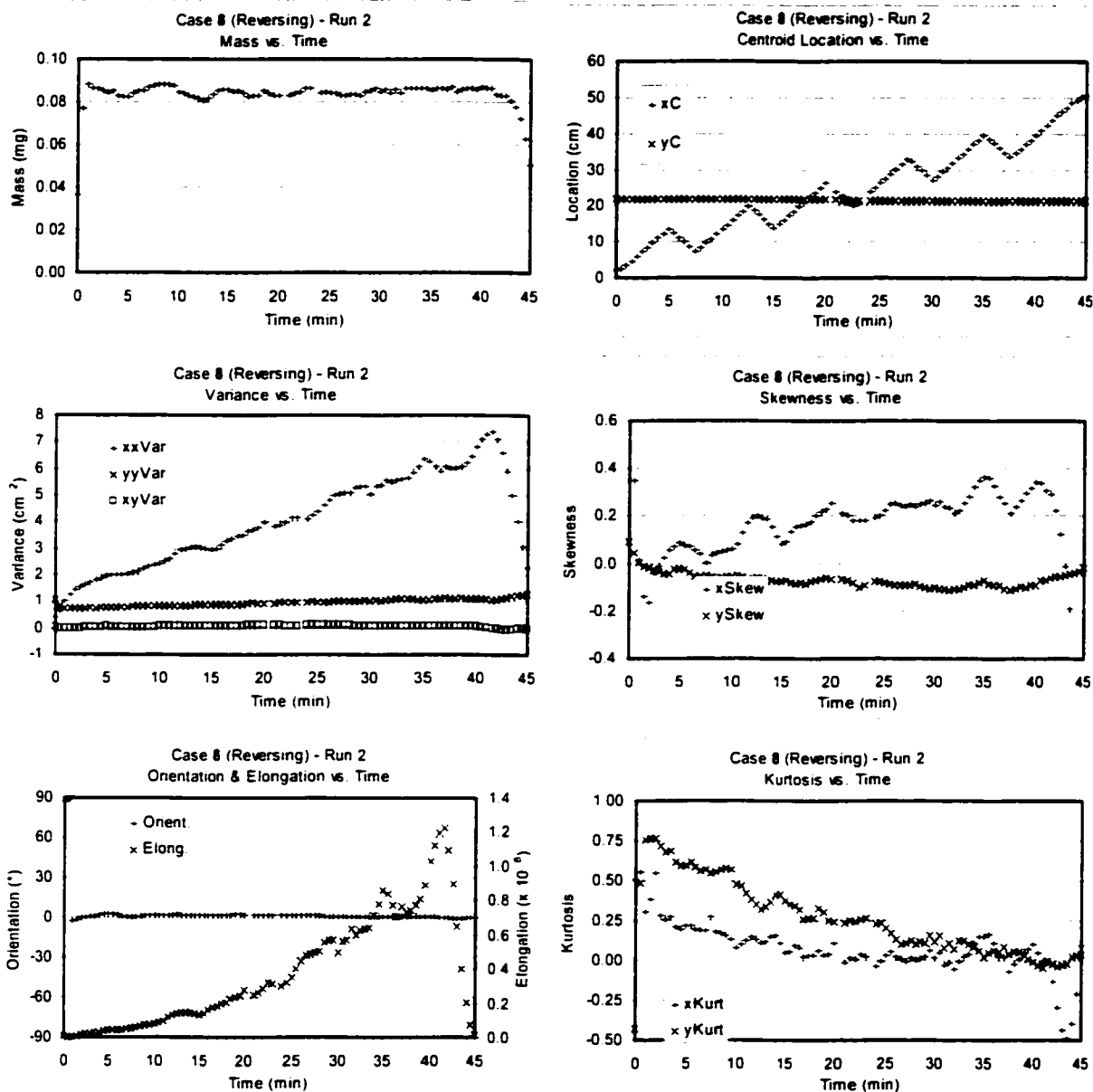


Case 8, run 1 (Reversing, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$
			cm min^{-1}	cm min^{-1}	cm min^{-1}	$\text{cm}^2 \text{min}^{-1}$	$\text{cm}^2 \text{min}^{-1}$	cm	cm	cm	cm
Method	1	79	1	0.858	-0.011	0.858	0.069	0.005	0.081	0.005	-
	2	7	11	0.973	-0.009	0.973	0.077	0.004	0.079	0.004	0.030
	3	7	11	2.530	0.012	2.530	0.077	0.004	0.030	0.002	0.030
	4	-	78	0.964	-0.010	0.964	0.076	0.004	0.079	0.004	0.019

Homogeneous Medium – Laboratory Results

Case 8, run 2 (Reversing, $i = 0.2174$)



Case 8, run 2 (Reversing, $i = 0.2174$)

	N	M	v_x	v_y	$\ v\ $	$\langle D_x \rangle$	$\langle D_y \rangle$	A_x	A_y	$\langle A_x \rangle$	$\langle A_y \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	77	1	0.859	-0.010	0.859	0.069	0.005	0.081	0.006	-	-
	2	7	11	0.968	-0.008	0.968	0.078	0.004	0.081	0.005	0.031	0.002
	3	7	11	2.504	0.012	2.504	0.078	0.004	0.031	0.002	0.031	0.002
	4	-	76	0.956	-0.009	0.956	0.075	0.004	0.079	0.004	0.021	0.002

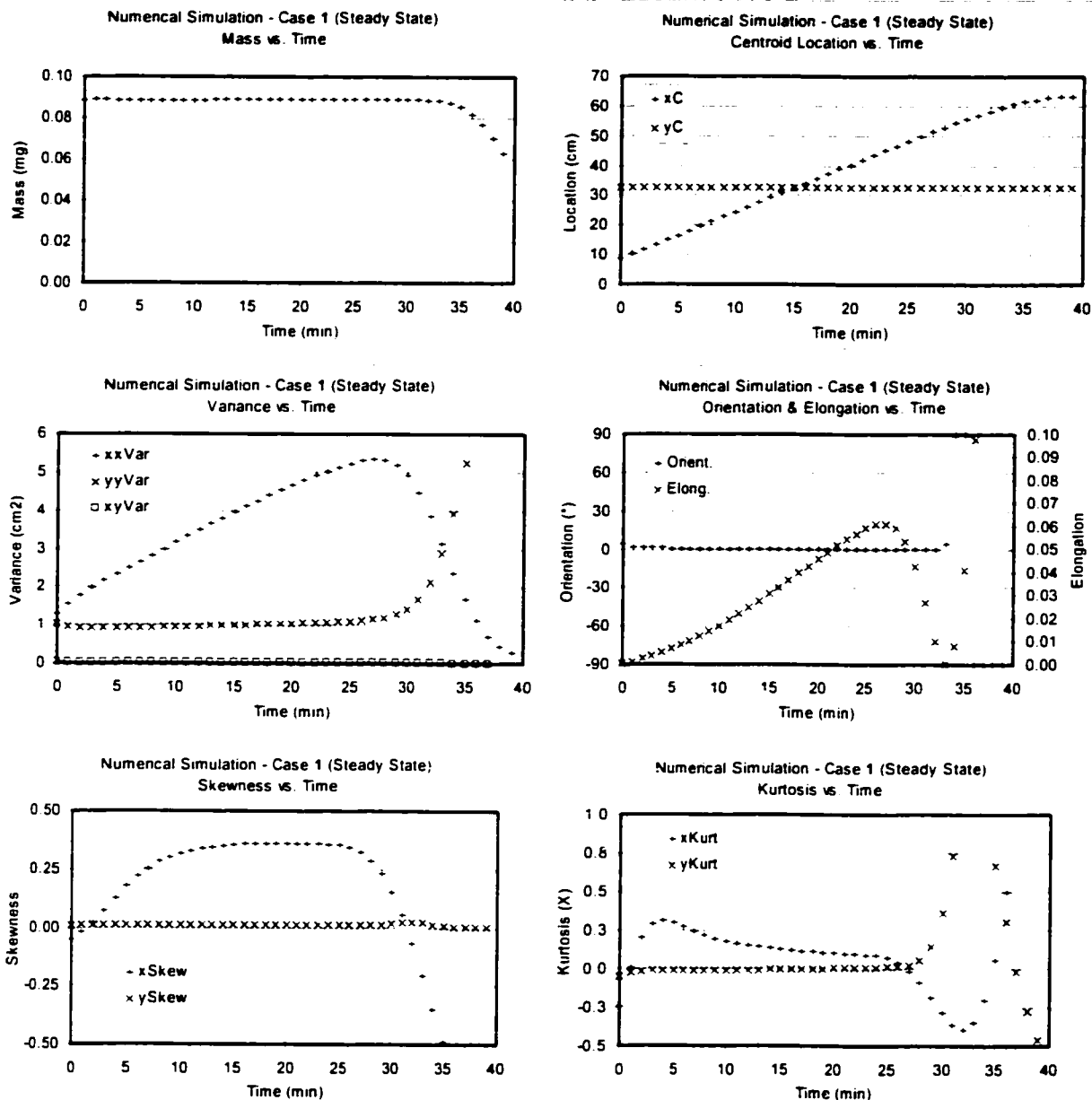
2.2 Numerical simulations

The following results obtained from the numerical simulations performed using a homogeneous porous medium (discussed in Chapter 3) are each shown below on a separate page:

1. Hydraulic gradient $i = 0.1211$:
 - a. Case 1: Steady state scenario
 - b. Case 2: Zigzag 30° scenario
 - c. Case 3: Zigzag 60° scenario
 - d. Case 4: Reversing scenario
2. Hydraulic gradient $i = 0.2174$:
 - a. Case 5: Steady state scenario
 - b. Case 6: Zigzag 30° scenario
 - c. Case 7: Zigzag 60° scenario
 - d. Case 8: Reversing scenario

Homogeneous Medium – Numerical Simulations

Case 1 (Steady State, $i = 0.1211$)

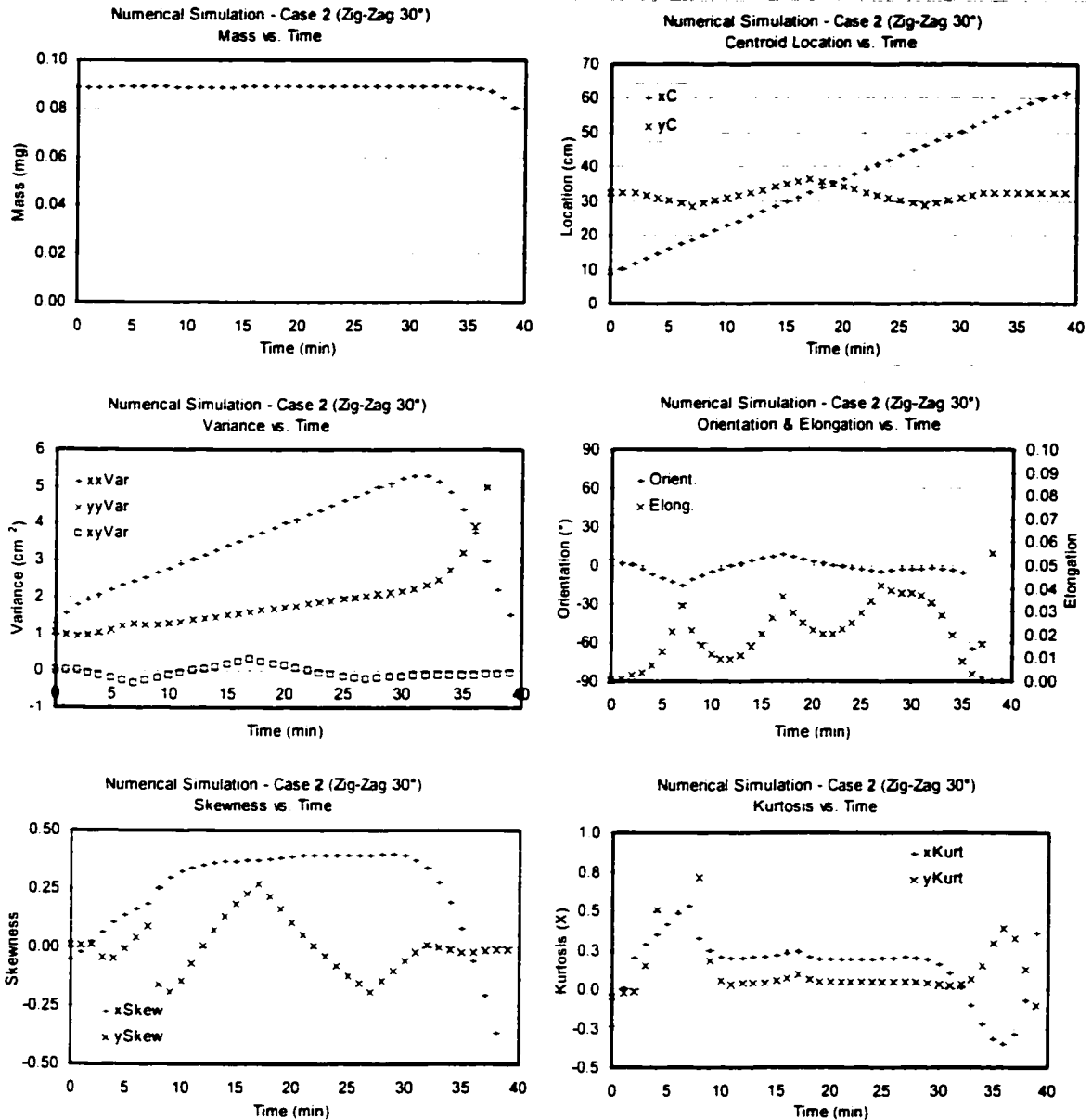


Case 1 (Homogeneous medium, Steady State flow, $i = 0.1211$)

	N	M	v_1	v_2	$ v $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	28	1	1.585	0.000	1.585	0.076	0.003	0.048	0.002	-
	2	28	1	1.585	0.000	1.585	0.076	0.003	0.048	0.002	0.048
	3	28	1	1.585	0.000	1.585	0.076	0.003	0.048	0.002	0.048
	4	-	27	1.575	0.000	1.575	0.075	0.002	0.048	0.001	0.048

Homogeneous Medium – Numerical Simulations

Case 2 (Zigzag 30°, $i = 0.1211$)

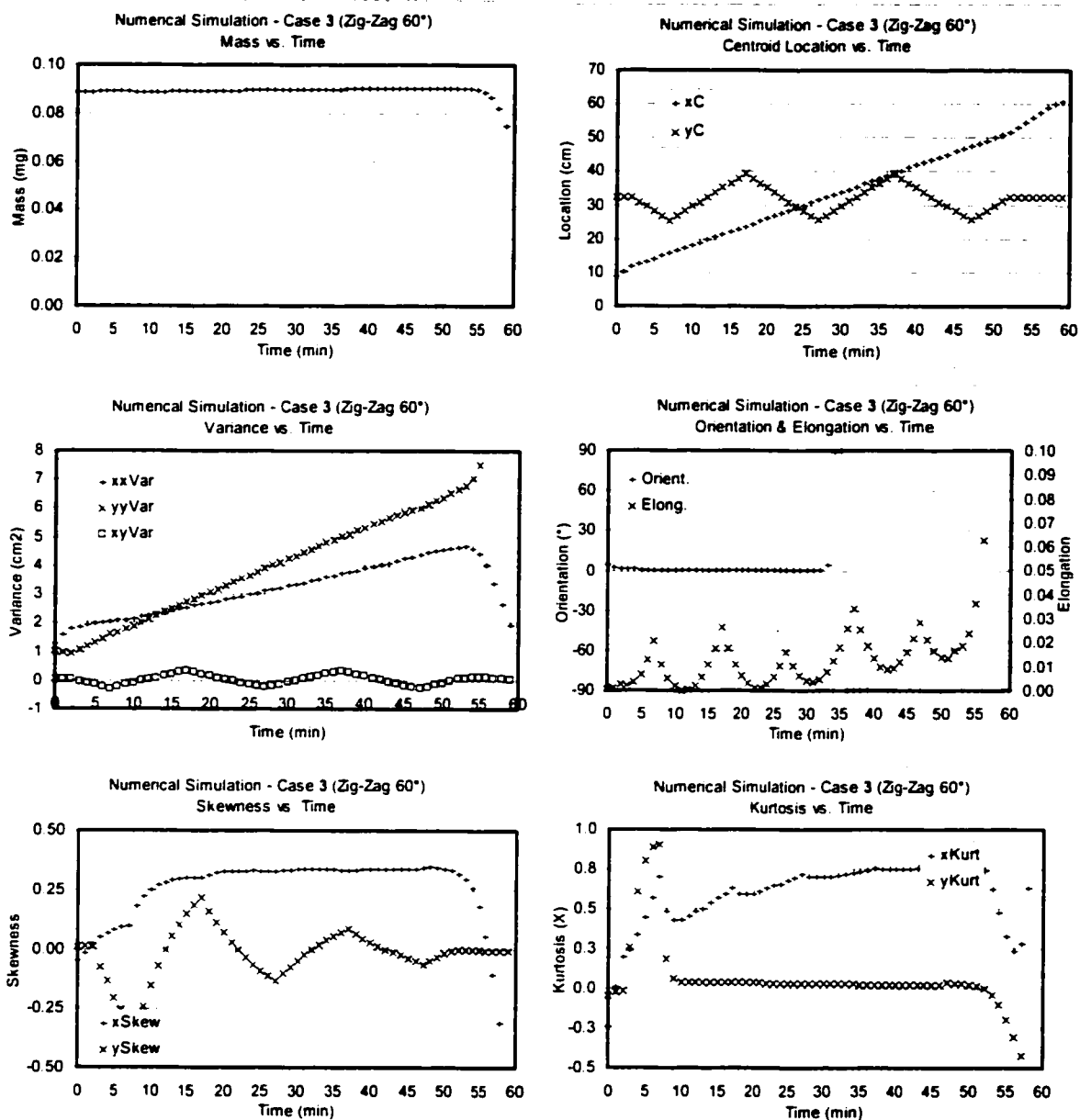


Case 2 (Homogeneous medium, Zig-Zag 30° flow pattern, $i = 0.1211$)

	N	M	v_1 cm min ⁻¹	v_2 cm min ⁻¹	$\ v\ $ cm min ⁻¹	$\langle D_{11} \rangle$ cm ² min ⁻¹	$\langle D_{22} \rangle$ cm ² min ⁻¹	A_{11} cm	A_{22} cm	$\langle A_{11} \rangle$ cm	$\langle A_{22} \rangle$ cm	
Method	1	32	1	1.378	-0.008	1.378	0.062	0.021	0.045	0.015	-	-
	2	7	5	1.384	-0.024	1.384	0.065	0.020	0.047	0.015	0.041	0.013
	3	7	5	1.384	0.739	1.569	0.065	0.020	0.041	0.013	0.041	0.013
	4	-	31	1.384	-0.025	1.384	0.064	0.019	0.047	0.014	0.041	0.012

Homogeneous Medium – Numerical Simulations

Case 3 (Zigzag 60°, $i = 0.1211$)

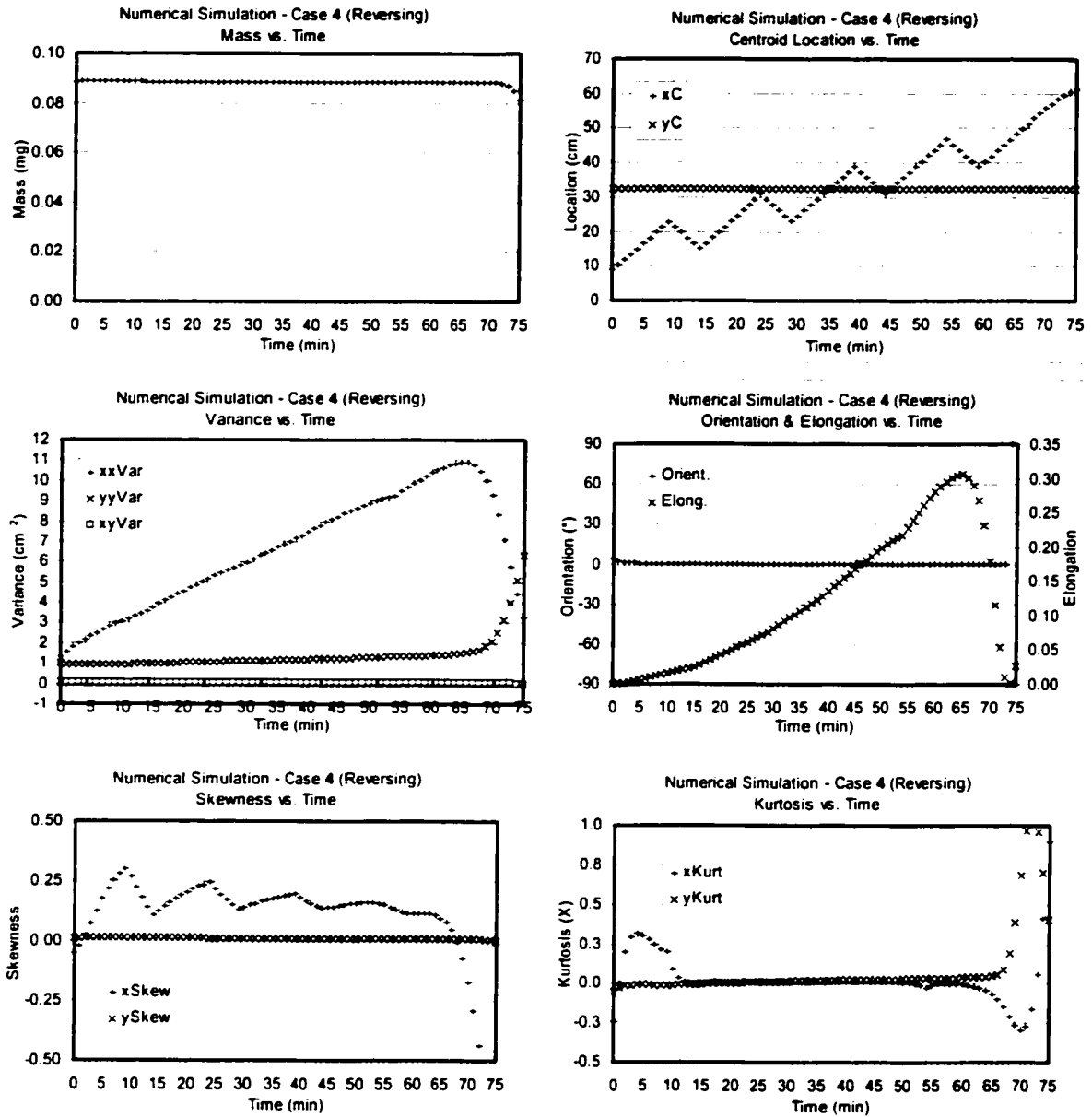


Case 3 (Homogeneous medium, Zig-Zag 60° flow pattern, $i = 0.1211$)

	N	M	v_1 cm min ⁻¹	v_2 cm min ⁻¹	$\ v\ $ cm min ⁻¹	$\langle D_{11} \rangle$ cm ² min ⁻¹	$\langle D_{22} \rangle$ cm ² min ⁻¹	A_{11} cm	A_{22} cm	$\langle A_{11} \rangle$ cm	$\langle A_{22} \rangle$ cm	
Method	1	53	1	0.798	-0.001	0.798	0.029	0.056	0.037	0.070	-	-
	2	8	7	0.821	0.001	0.821	0.032	0.054	0.039	0.066	0.020	0.034
	3	8	7	0.821	1.327	1.561	0.032	0.054	0.021	0.035	0.020	0.034
	4	-	52	0.822	0.001	0.822	0.032	0.054	0.039	0.066	0.020	0.034

Homogeneous Medium – Numerical Simulations

Case 4 (Reversing, $i = 0.1211$)

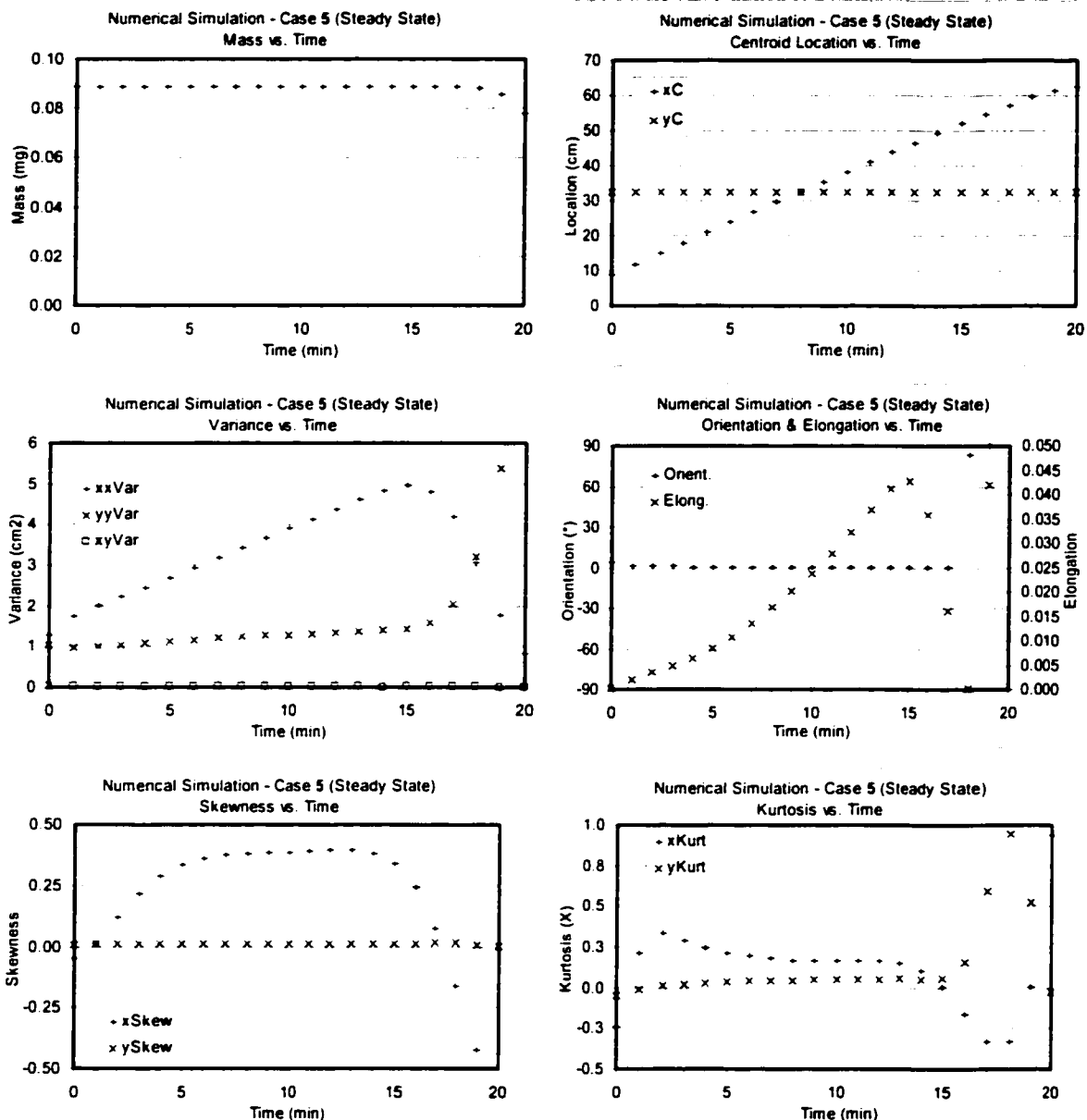


Case 4 (Homogeneous medium, Reversing flow pattern, $i = 0.1211$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	66	1	0.522	0.000	0.522	0.073	0.004	0.140	0.008	–
	2	8	9	0.607	0.000	0.607	0.074	0.004	0.121	0.006	0.047
	3	8	9	1.585	0.000	1.585	0.074	0.004	0.047	0.002	0.047
	4	–	65	0.607	0.000	0.607	0.074	0.004	0.122	0.006	0.047

Homogeneous Medium – Numerical Simulations

Case 5 (Steady State, $i = 0.2174$)

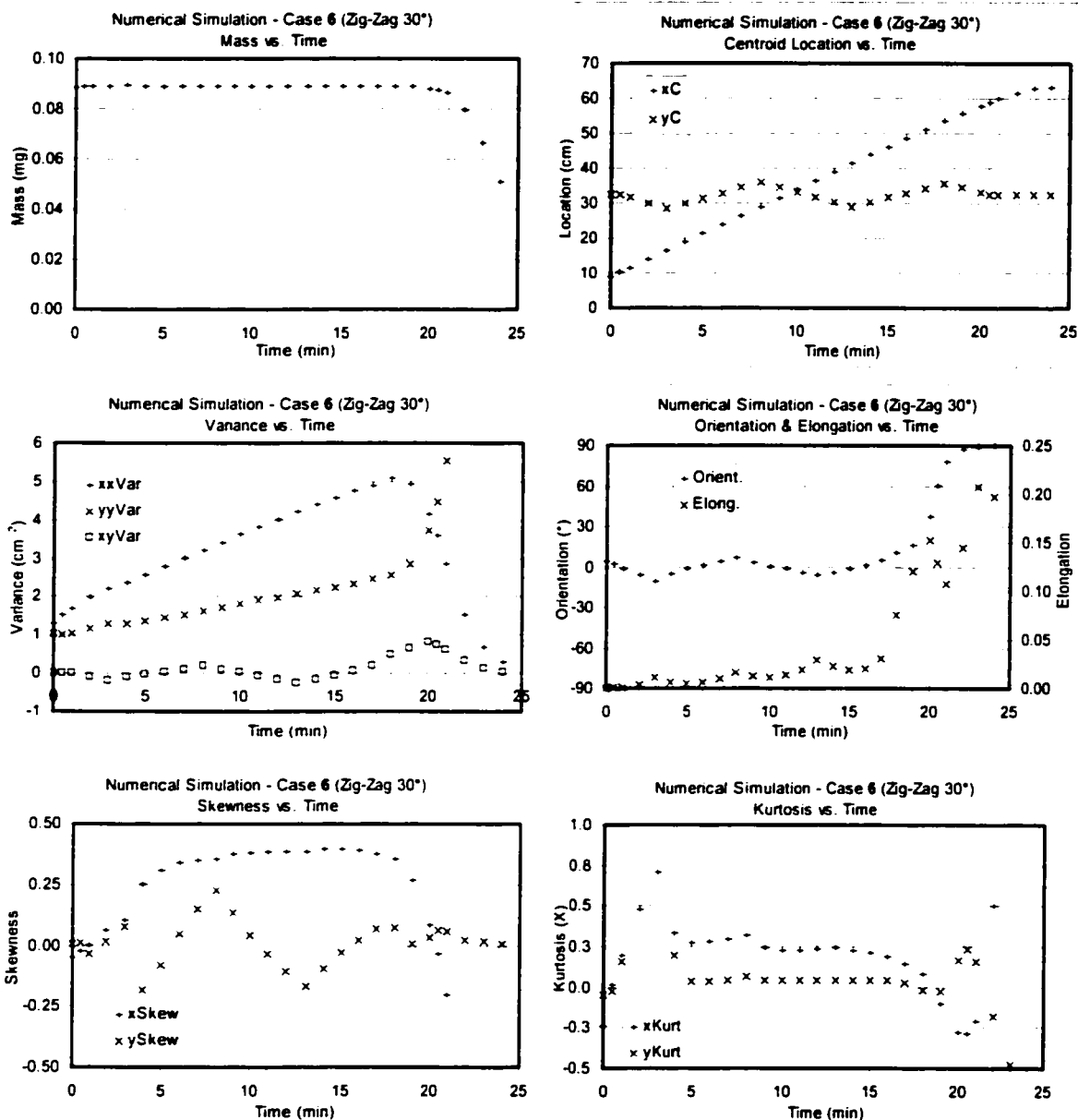


Case 5 (Homogeneous medium, Steady State flow, $i = 0.2174$)

		N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
				cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	16	1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	-	-
	2	16	1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	0.042	0.005
	3	16	1	2.880	0.000	2.880	0.120	0.015	0.042	0.005	0.042	0.005
	4	-	15	2.884	0.000	2.884	0.122	0.014	0.042	0.005	0.042	0.005

Homogeneous Medium - Numerical Simulations

Case 6 (Zigzag 30°, $i = 0.2174$)

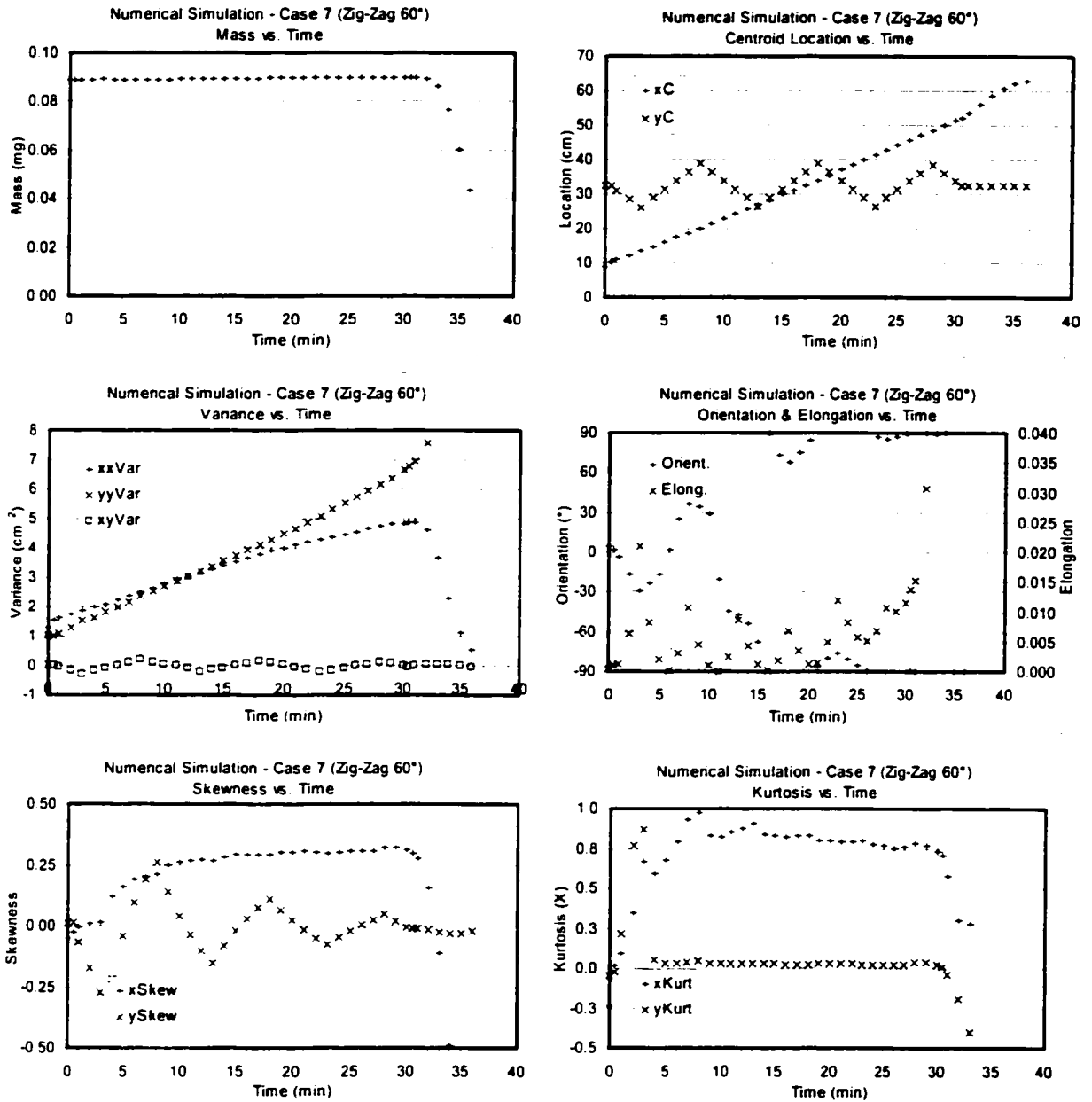


Case 6 (Homogeneous medium, Zig-Zag 30° flow pattern, $i = 0.2174$)

	N	M	v_1	v_2	$ v $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	21	1	2.474	0.118	2.476	0.099	0.045	0.040	0.018	-	-
	2	4	6	2.481	0.185	2.488	0.106	0.043	0.042	0.017	0.037	0.015
	3	4	6	2.481	1.428	2.862	0.106	0.043	0.037	0.015	0.037	0.015
	4	-	20	2.474	0.108	2.477	0.096	0.048	0.039	0.019	0.033	0.017

Homogeneous Medium – Numerical Simulations

Case 7 (Zigzag 60°, $i = 0.2174$)

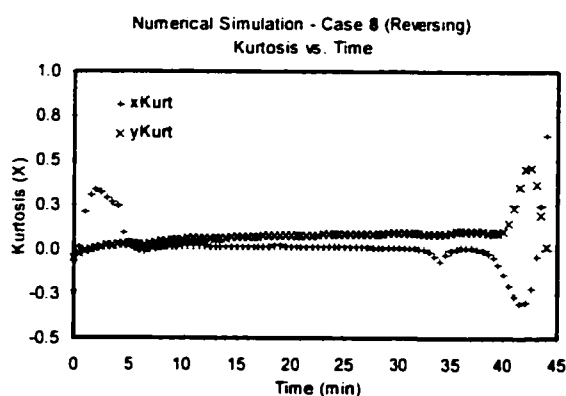
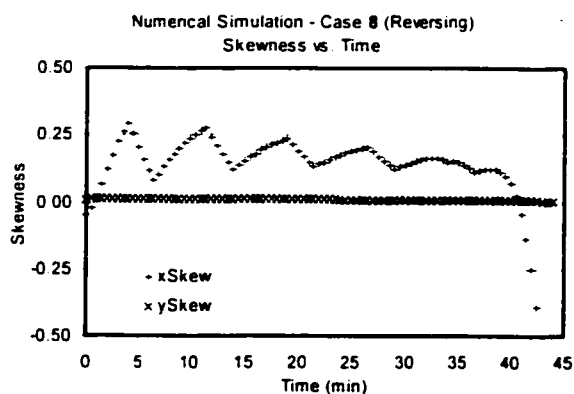
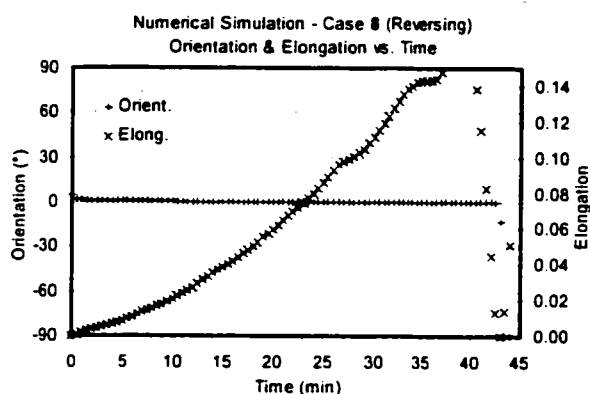
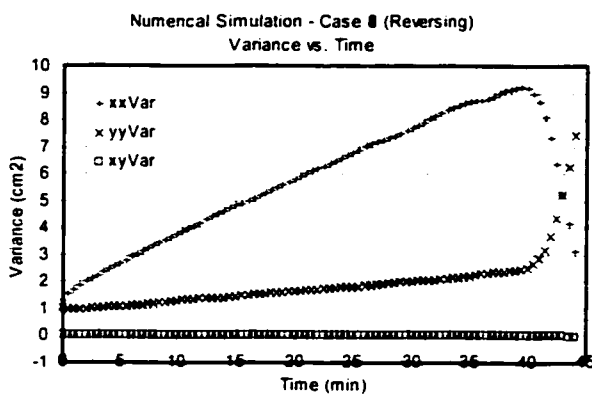
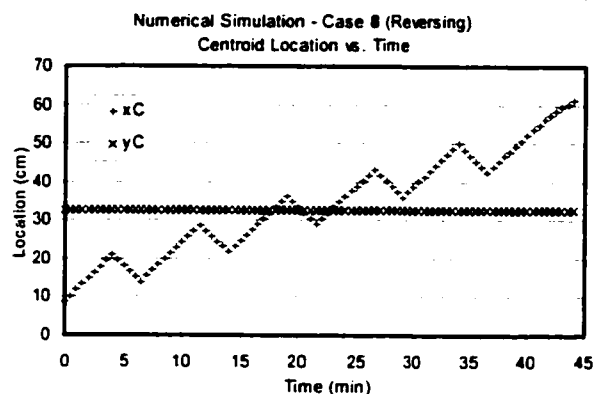
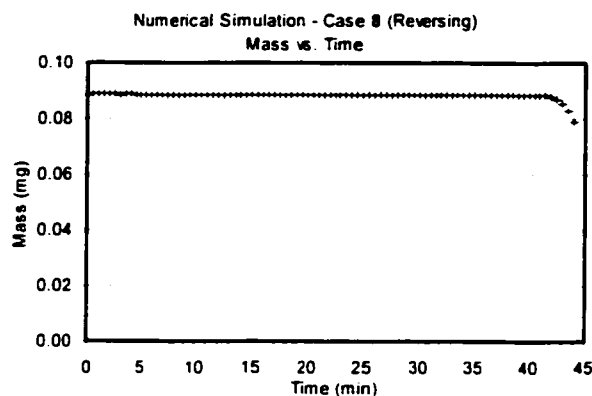


Case 7 (Homogeneous medium, Zig-Zag 60° flow pattern, $i = 0.2174$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	33	1	1.404	0.084	1.406	0.058	0.093	0.041	0.066	-	-
	2	5	8	1.417	0.005	1.417	0.059	0.095	0.041	0.067	0.021	0.033
	3	5	8	1.417	2.442	2.824	0.059	0.095	0.021	0.034	0.021	0.033
	4	-	32	1.418	0.005	1.418	0.058	0.095	0.041	0.067	0.020	0.033

Homogeneous Medium – Numerical Simulations

Case 8 (Reversing, $i = 0.2174$)



Case 8 (Homogeneous medium, Reversing flow pattern, $i = 0.2174$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	81	1	0.949	0.000	0.949	0.099	0.019	0.104	0.020	-
	2	8	11	1.083	0.000	1.083	0.099	0.019	0.091	0.017	0.034
	3	8	11	2.885	0.001	2.885	0.099	0.019	0.034	0.007	0.034
	4	-	80	1.083	0.000	1.083	0.098	0.019	0.091	0.017	0.000

3 RESULTS FROM CHAPTER 4 – HETEROGENEOUS POROUS MEDIUM

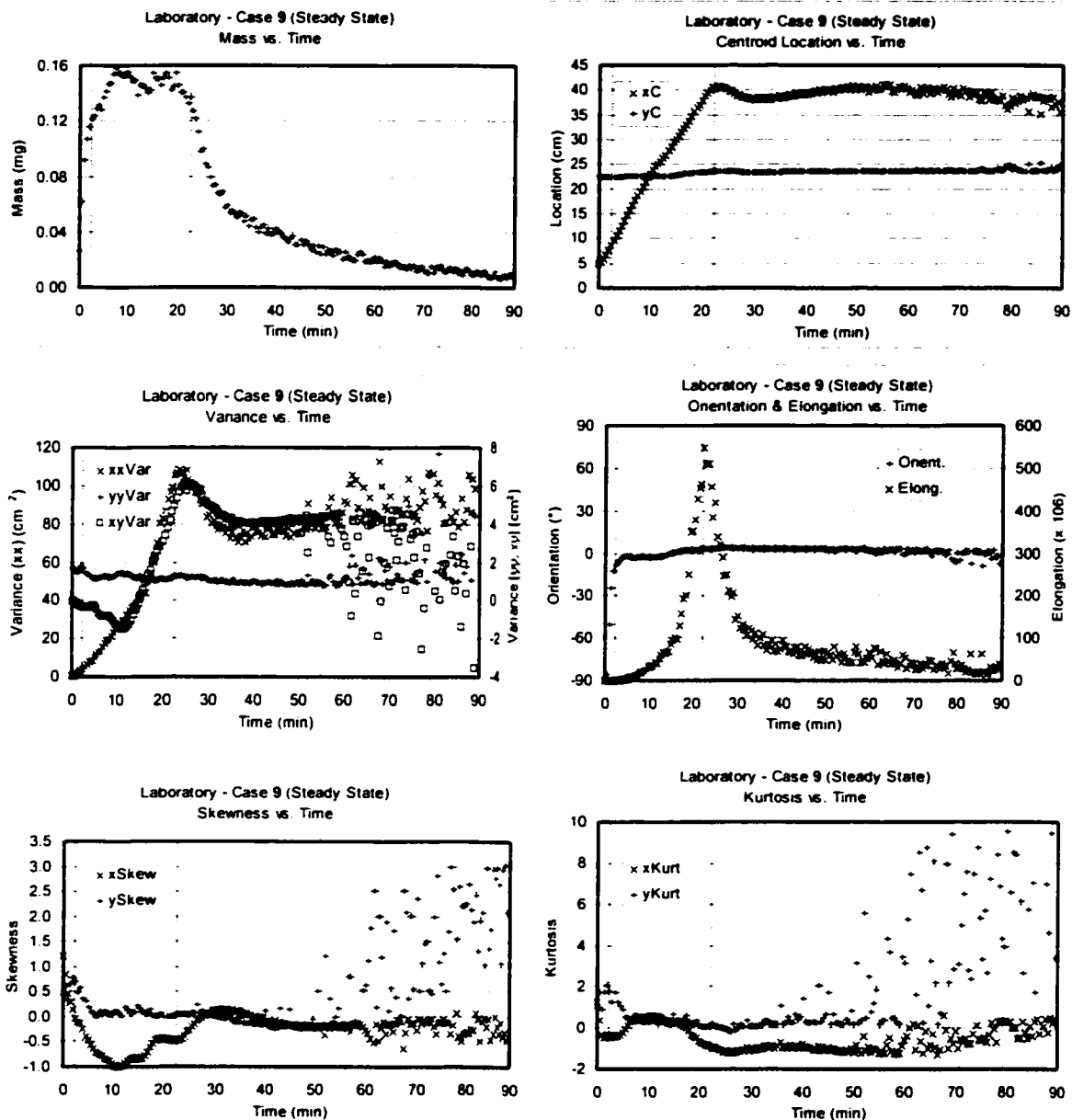
3.1 Laboratory experiments

The following results obtained from the laboratory experiments performed using a heterogeneous porous medium (discussed in Chapter 4) are shown below (η represents the number of replicate runs, each shown on a separate page):

1. Hydraulic gradient $i = 0.2133$:
 - a. Case 9: Steady state scenario ($\eta = 2$)
 - b. Case 10: Zigzag 30° scenario ($\eta = 2$)
 - c. Case 11: Zigzag 60° scenario ($\eta = 2$)
 - d. Case 12: Reversing scenario ($\eta = 2$)

Heterogeneous Medium - Laboratory Experiments

Case 9 (Steady State, $i = 0.2133$)

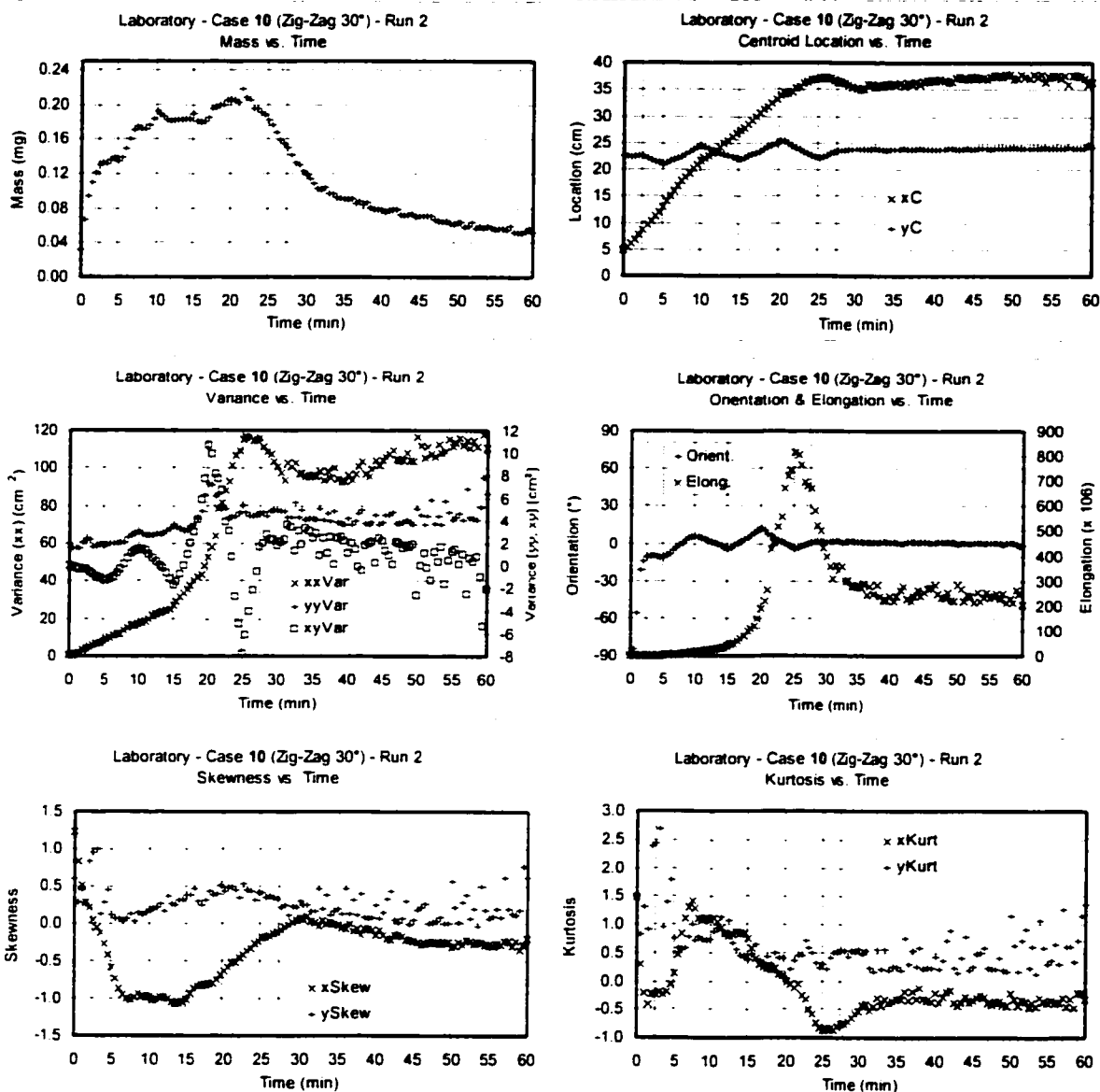


Case 9 (Heterogeneous, Steady State, $i = 0.2133$)

Method		N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
				cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
	1	41	1	1.591	0.058	1.592	2.322	-0.006	1.459	-0.004	-	-
	2	41	1	1.591	0.058	1.592	2.322	-0.006	1.459	-0.004	1.459	-0.004
	3	41	1	1.591	0.058	1.592	2.322	-0.006	1.459	-0.004	1.459	-0.004
	4	-	40	1.588	0.058	1.589	2.552	-0.010	1.606	-0.006	2.616	0.015

Heterogeneous Medium – Laboratory Experiments

Case 10 (Zigzag 30°, $i = 0.2133$)

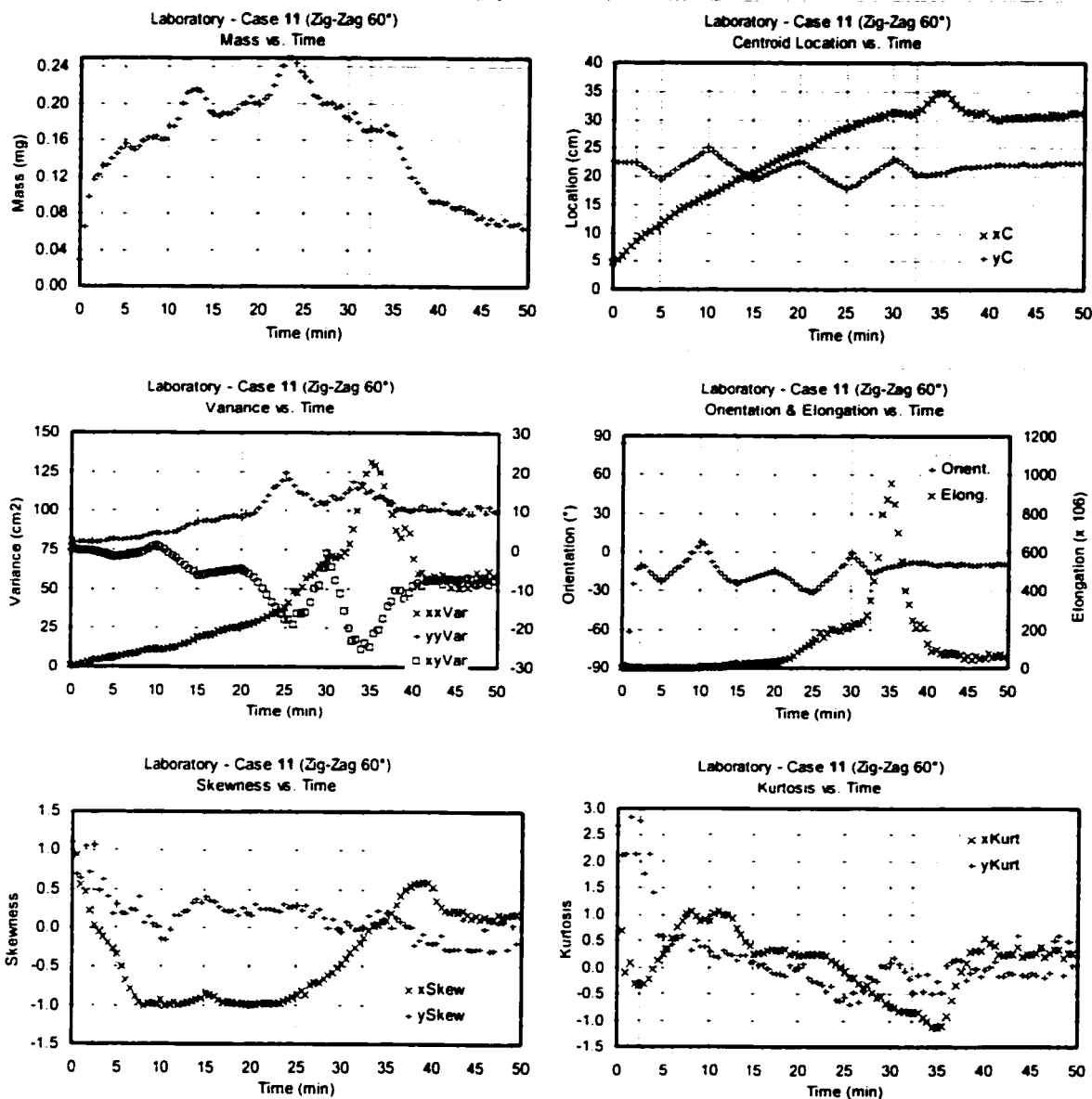


Case 10 (Heterogeneous, Zig-Zag 30°, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	50	1	1.179	0.060	1.181	1.649	0.108	1.397	0.091	—
	2	9	6	1.119	0.038	1.119	2.212	0.027	1.976	0.024	1.848
	3	9	6	1.183	0.661	1.355	2.212	0.027	1.633	0.020	1.848
	4	—	49	1.106	0.040	1.107	2.232	0.046	2.016	0.041	1.818

Heterogeneous Medium – Laboratory Experiments

Case 11 (Zigzag 60°, $i = 0.2133$)

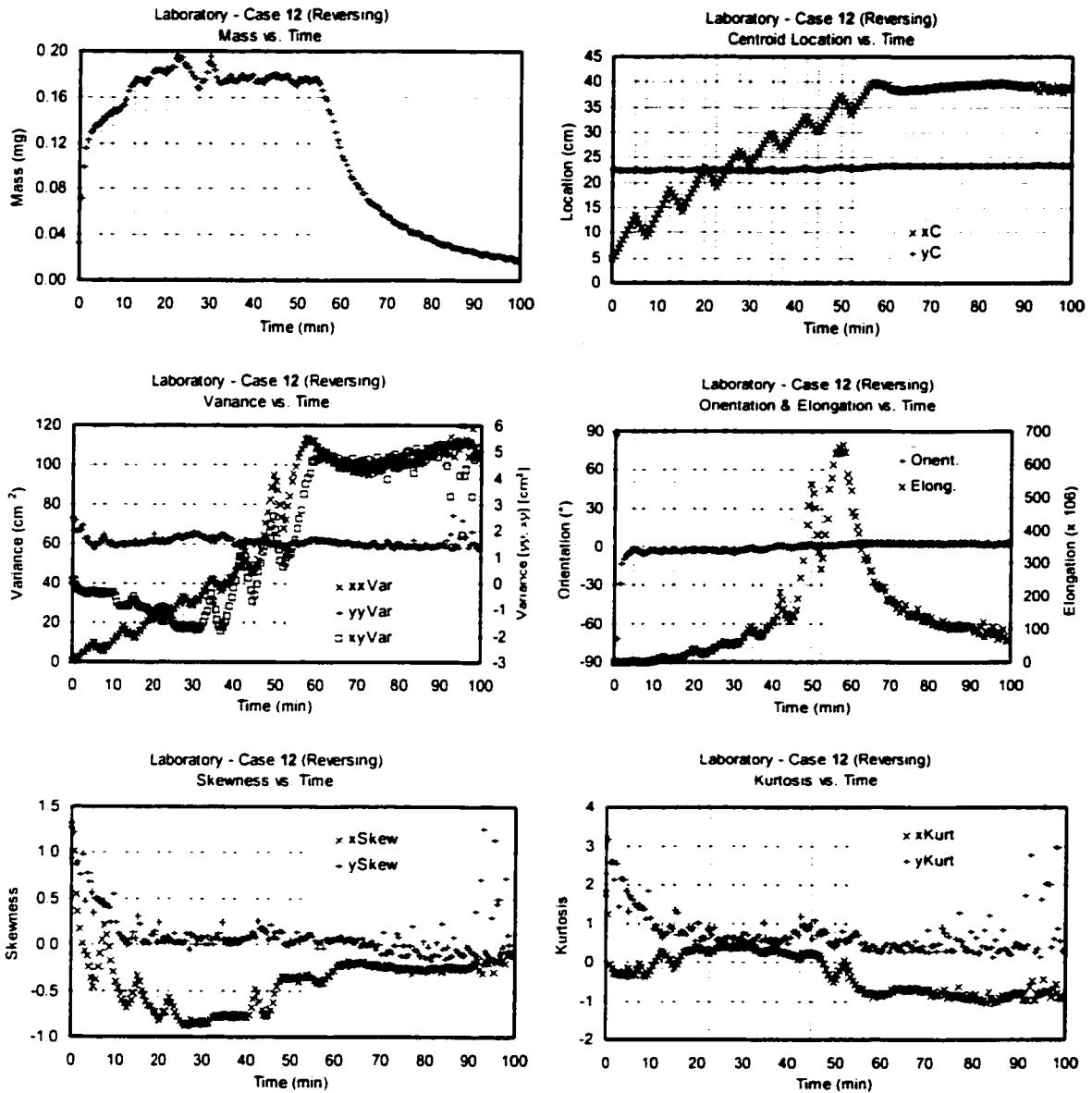


Case 11 (Heterogeneous, Zig-Zag 60°, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	64	1	0.762	-0.048	0.764	1.373	0.253	1.797	0.331	-	-
	2	8	8	0.826	-0.086	0.830	1.957	0.221	2.358	0.266	1.476	0.161
	3	8	8	0.826	0.963	1.269	1.957	0.221	1.543	0.174	1.476	0.161
	4	-	63	0.813	-0.067	0.815	1.871	0.189	2.295	0.232	1.474	0.181

Heterogeneous Medium – Laboratory Experiments

Case 12 (Reversing, $i = 0.2133$)



Case 12 (Heterogeneous, Reversing, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	101	1	0.530	0.008	0.530	0.708	0.000	1.334	0.000	-
	2	8	14	0.540	0.010	0.540	0.758	-0.007	1.404	-0.014	0.438
	3	8	14	1.465	0.058	1.466	0.758	-0.007	0.517	-0.005	0.438
	4	-	100	0.514	0.006	0.514	0.682	-0.008	1.327	-0.015	0.194

3.2 Numerical simulations

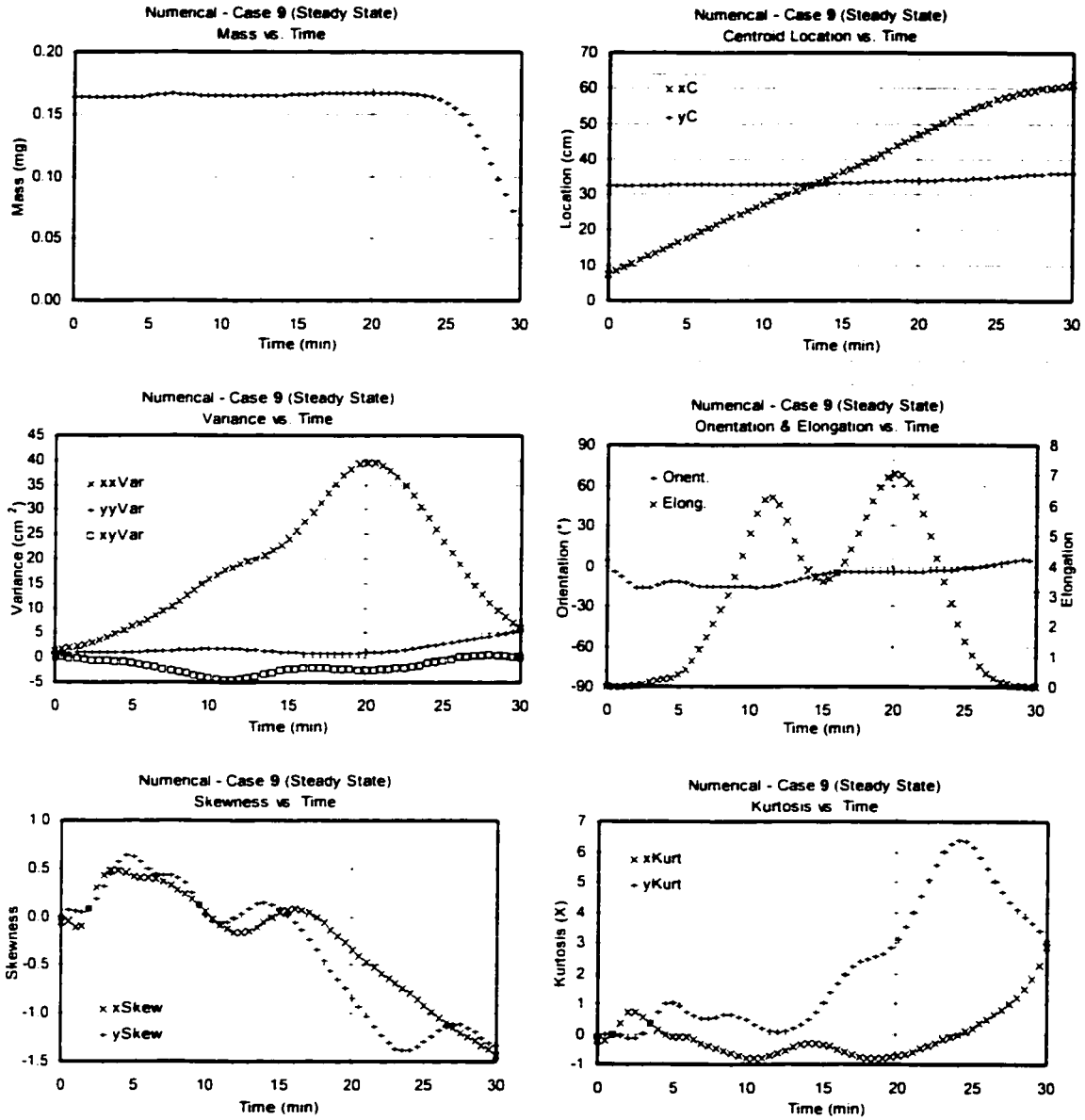
The following results obtained from the numerical simulations performed using a heterogeneous porous medium (discussed in Chapter 4) are each shown below on a separate page:

2. Hydraulic gradient $i = 0.2133$:

- a. Case 9: Steady state scenario ($\eta = 2$)
- b. Case 10: Zigzag 30° scenario ($\eta = 2$)
- c. Case 11: Zigzag 60° scenario ($\eta = 2$)
- d. Case 12: Reversing scenario ($\eta = 2$)

Heterogeneous Medium - Numerical Simulations

Case 9 (Steady State, $i = 0.2133$)

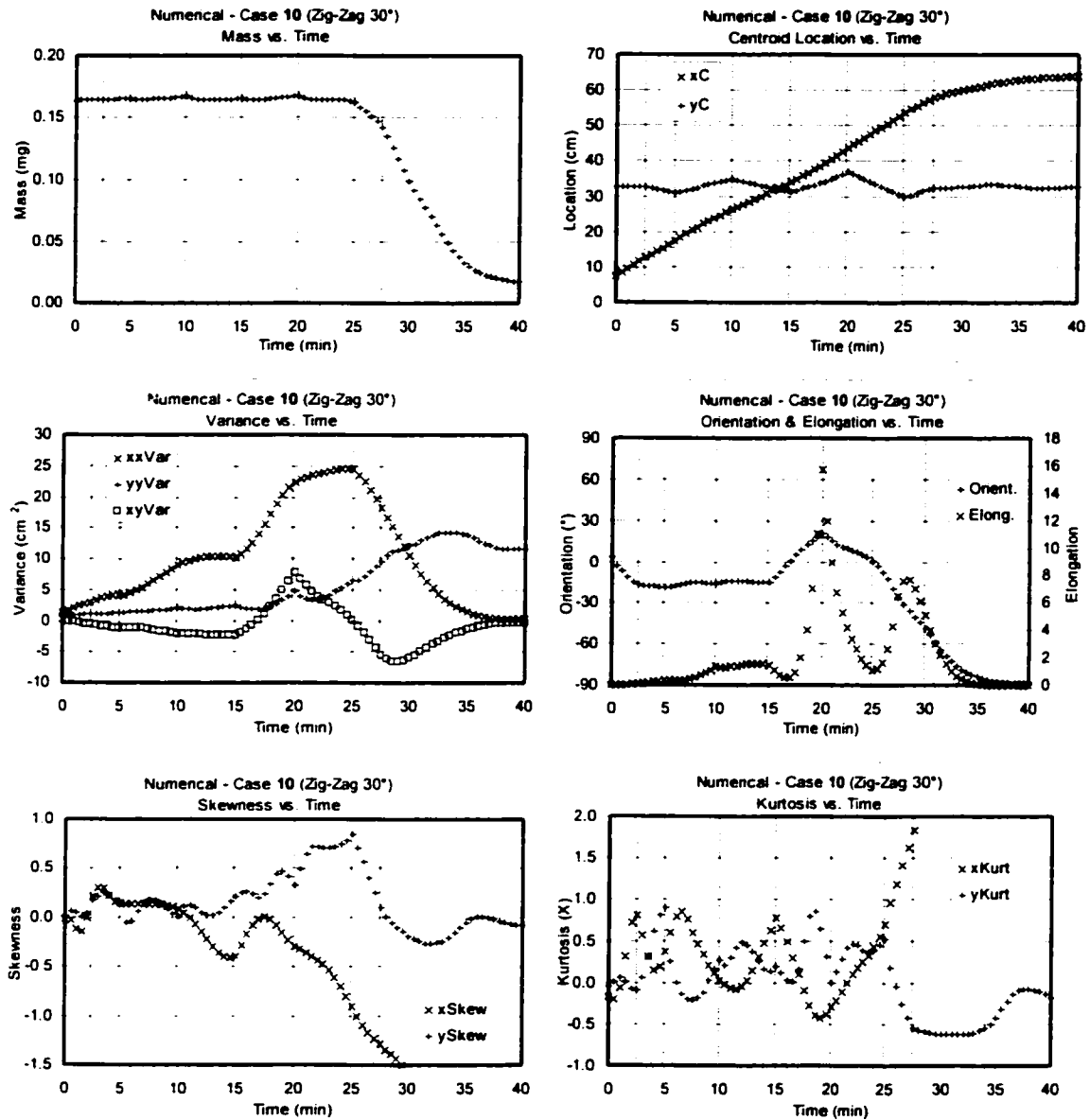


Case 9 (Heterogeneous, Steady State, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	41	1	1.937	0.059	1.938	0.973	-0.003	0.502	-0.002	-
	2	41	1	1.937	0.059	1.938	0.973	-0.003	0.502	-0.002	0.502
	3	41	1	1.937	0.059	1.938	0.973	-0.003	0.502	-0.002	0.502
	4	-	40	1.987	0.071	1.988	0.957	-0.002	0.481	-0.001	0.479

Heterogeneous Medium – Numerical Simulations

Case 10 (Zigzag 30°, $i = 0.2133$)

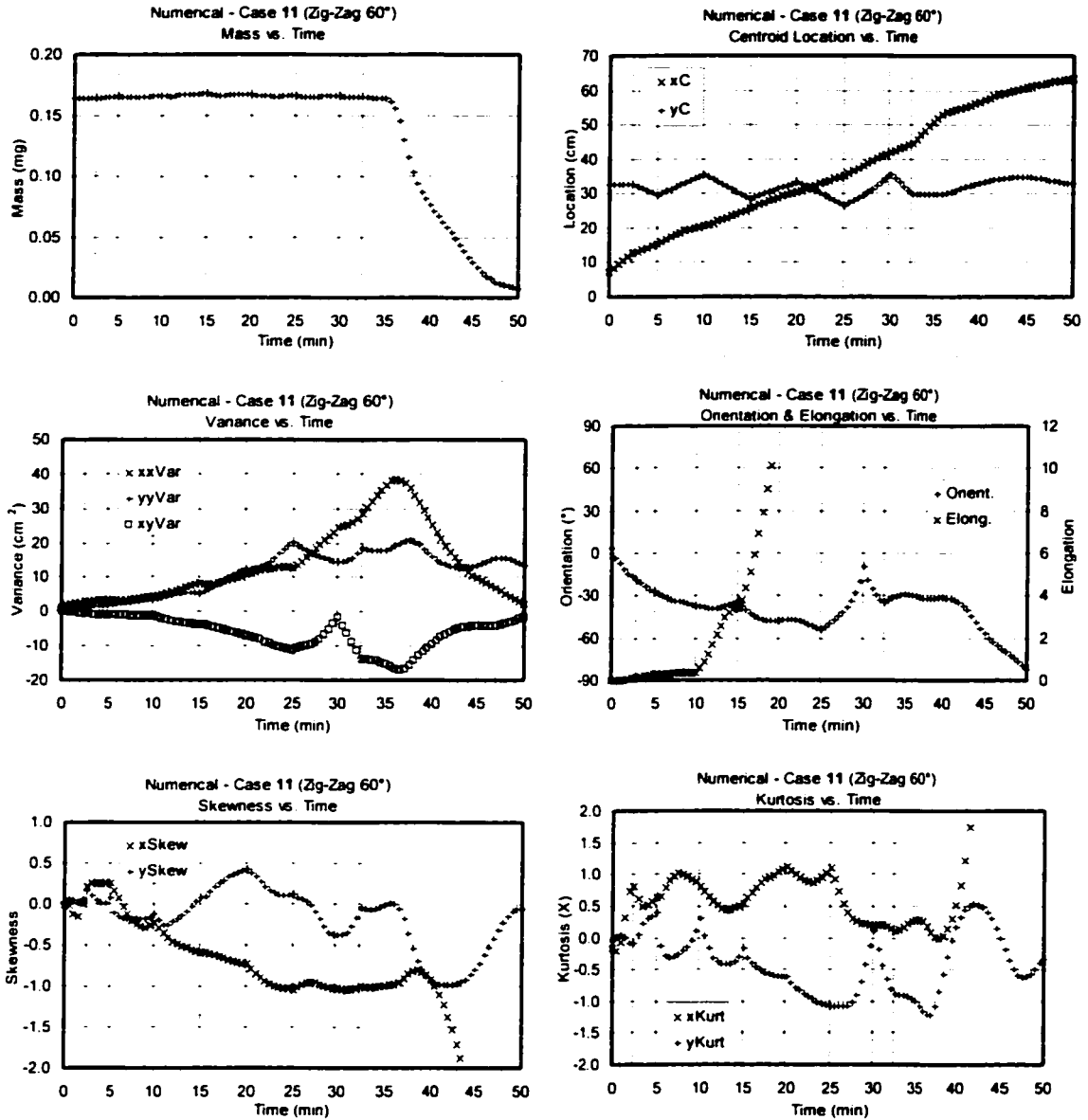


Case 10 (Heterogeneous, Zig-Zag 30°, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	27	1	1.781	0.049	1.782	0.512	0.079	0.287	0.044	–
	2	9	6	1.855	-0.113	1.859	0.491	0.124	0.264	0.067	0.232
	3	9	6	1.855	0.857	2.044	0.491	0.124	0.240	0.061	0.232
	4	–	26	1.856	-0.111	1.859	0.467	0.110	0.251	0.059	0.098

Heterogeneous Medium – Numerical Simulations

Case 11 (Zigzag 60°, $i = 0.2133$)

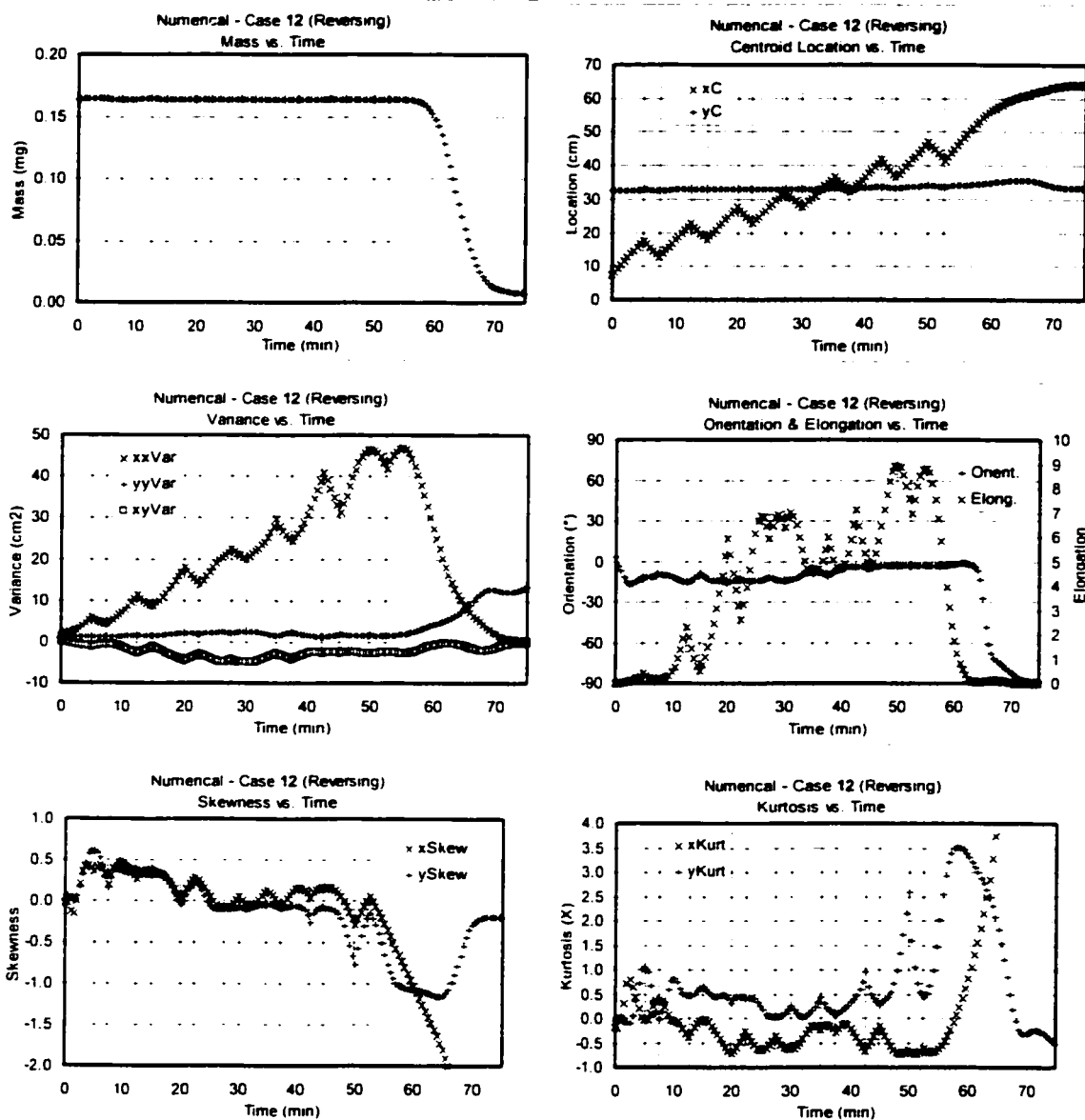


Case 11 (Heterogeneous, Zig-Zag 60°, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$	
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm	
Method	1	39	1	1.089	-0.056	1.090	0.418	0.300	0.384	0.275	-	-
	2	9	9	1.247	-0.090	1.250	0.523	0.262	0.418	0.209	0.253	0.172
	3	9	9	1.247	1.237	1.756	0.523	0.262	0.298	0.149	0.253	0.172
	4	-	38	1.241	-0.077	1.243	0.508	0.240	0.409	0.193	0.072	0.097

Heterogeneous Medium - Numerical Simulations

Case 12 (Reversing, $i = 0.2133$)



Case 12 (Heterogeneous, Reversing, $i = 0.2133$)

	N	M	v_1	v_2	$\ v\ $	$\langle D_{11} \rangle$	$\langle D_{22} \rangle$	A_{11}	A_{22}	$\langle A_{11} \rangle$	$\langle A_{22} \rangle$
			cm min ⁻¹	cm min ⁻¹	cm min ⁻¹	cm ² min ⁻¹	cm ² min ⁻¹	cm	cm	cm	cm
Method	1	89	1	0.641	0.022	0.642	0.423	0.003	0.659	0.005	-
	2	8	15	0.724	0.027	0.724	0.425	0.007	0.587	0.010	0.215
	3	8	15	1.974	0.079	1.976	0.425	0.007	0.215	0.004	0.215
	4	-	88	0.724	0.028	0.724	0.415	0.006	0.573	0.009	0.000