**What Have We Learned From Deterministic Geostatistics at Highly Resolved Field Sites,
as Relevant to Mass Transport Processes in Sedimentary Aquifers?**

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Abstract:

In the method of deterministic geostatistics (*sensu* Issaks and Srivastava, 1988), highly-resolved data sets are used to compute sample spatial-bivariate statistics within a deterministic framework. The general goal is to observe what real, highly resolved, sample spatial-bivariate correlation looks like when it is well-quantified in naturally-occurring sedimentary aquifers. Furthermore, it is to understand how this correlation structure, (i.e. shape and correlation range) is related to independent and physically quantifiable attributes of the sedimentary architecture. The approach has evolved among work by Rubin (1995), Barrash and Clemo (2002), Rubin

(2003), Ritzi et al. (2004); Dai et al. (2005), Ramanathan et al. (2010) and Ritzi et al. (2007, 2013). In this evolution, equations for sample statistics have been developed which allow tracking the facies types at the heads and tails of lag vectors. The goal is to observe and thereby understand how aspects of the sedimentary architecture affect the well-supported sample statistics. The approach has been used to study heterogeneity at a number of sites, representing a variety of depositional environments, with highly resolved data sets. What have we learned? We offer and support an opinion that the single most important insight derived from these studies is that the structure of spatial-bivariate correlation is essentially the cross-transition probability structure, determined by the sedimentary architecture. More than one scale of hierarchical sedimentary architecture has been represented in these studies, and a hierarchy of cross-transition probability structures was found to define the correlation structure in all cases. This insight allows decomposing contributions from different scales of the sedimentary architecture, and has led to a more fundamental understanding of mass transport processes including mechanical dispersion of solutes within aquifers, and the time-dependent retardation of reactive solutes. These processes can now be described in terms of quantifiable physical attributes of the sedimentary architecture. In addition to discussing the benefits to basic scientific understanding, we also discuss some of the practical benefits derived from this insight.

1. Introduction: Legacy Research Sites, and Evolution of Deterministic Geostatistics

During the 1980s, a number of highly-instrumented field-research sites were established for the study of mass transport processes; processes including advection, mechanical dispersion,

sorption, and biogeochemical transformation (see review by Anderson and McCray, 2011). The development of these dedicated field-research sites was motivated by a growing concern over contamination of groundwater, and a need to validate theories for the processes governing contaminant transport in the subsurface. The theories were theretofore largely untested at the field scale because of a number of factors including 1) the difficulty in accessing the subsurface and observing processes insitu, 2) previously existing contaminant plumes were from unquantified sources and located at sites that had been subject to uncontrolled and unquantified influences of geologic, hydrologic and climatic factors, 3) developing dedicated field experiments with controlled, original releases requires a long time frame, and 4) the costs for infrastructure, operation and maintenance of field-scale experiments are very high (Sudicky and Illman, 2011). There were already enough data to suggest that field-scale processes could not be understood from lab-scale experiments alone. Thus, it was clear that a large investment was needed to create dedicated field research sites and thereby address the basic-science need for understanding mass transport processes in the subsurface. A basic-science understanding would require controlled releases of mass into a highly-characterized aquifer, with vast numbers of spatially distributed monitoring points in the subsurface, far more than would ever be installed at a typical industrial-contamination project site, in order to quantify, with highly-resolved data, the spatial distribution of the relevant attributes (e.g. permeability) and to quantify the spatio-temporal distribution of mass concentrations.

The first created was the Borden research site (see review by Sudicky and Illman, 2011). Following that were sites at Cape Cod, and the MADE site, and others (Anderson and McCay, 2011). Importantly, these sites were developed in unconsolidated sedimentary aquifers, reflecting the fact that nationally most groundwater withdrawal, and consequently most concern

over groundwater quality, is in such aquifers. The Borden site was chosen, in part, because the aquifer, a lake-shoreline sand deposit, is one of the more homogeneous sand aquifers one might encounter in nature, and there was a desire for hydrogeologic simplicity to avoid excessive uncertainty (Sudicky and Illman, 2011). One goal of the natural-gradient tracer experiments conducted at the Borden and Cape Cod sites was to test a Lagrangian-based stochastic theory for the dispersion of conservative solutes (Sudicky, 1986; Hess et al., 1992). This theory describes dispersion due to spatial variability in permeability, and permeability is geostatistically represented in the theory through its spatial-bivariate correlation. The original theory is appropriate in the context of explaining dispersion within a single aquifer stratum with mild heterogeneity, i.e. where the correlation range is small compared to the scale of plume migration, and the variance is mild and can be assumed as second-order stationary (Gelhar and Axness, 1983; Dagan, 1989). The theory has since been expanded (e.g. de Barros et al., 2015), including an expansion to explain reactive solute transport discussed below. The Borden and Cape Cod sites were appropriate for testing the early developments of this theory. The MADE site was not, as the tracer test comprised both aquifer and aquitard regions, and thus the MADE site is not discussed hereafter. Furthermore, the full spectrum of the research conducted among legacy research sites is much broader than simply testing the Lagrangian-based transport theory, and is not reviewed here.

The Lagrangian-based stochastic theory is built on a geostatistical foundation. Spatial-bivariate statistics are used to represent the intrinsic natural variability of permeability. The correlation range is the critical parameter. A goal of the early work at the Borden and Cape Cod sites was to deterministically quantify the spatial correlation in permeability along the path sampled by a tracer plume. Highly-resolved permeability data sets were developed to rigorously

quantify the sample spatial-bivariate correlation. We refer to this as deterministic geostatistics (*sensu* Issaks and Srivastava, 1988), as distinguished from other areas of geostatistics that focus on estimation and uncertainty. Only deterministic applications of geostatistics, within this context, are reviewed in this article.

In these early studies, the focus was almost entirely on the spatial variation of permeability itself, without much attention given to the underlying sedimentary architecture giving rise to the intrinsic natural variability in permeability. While cursory descriptions of the sedimentary architecture were given, there were no formal attempts to quantitatively characterize attributes of the sedimentary architecture in ways that could be directly included within the correlation models or the transport models (Sudicky, 1986; Woodbury and Sudicky, 1991; Hess et al., 1992).

This article reviews developments since that time, with a focus on how deterministic applications of geostatistics have evolved to include a formal and quantitative representation of the sedimentary architecture. We review how sample spatial-bivariate statistics have been formulated to incorporate representations of multi-scaled and hierarchical sedimentary architecture. We also review how these sample statistics have been used to understand heterogeneity at sites with highly resolved data, among different depositional environments. The main goal is to address the question of what we have learned from this approach. After a review of the relevant literature, we offer and support an opinion for what is the single most important insight derived from this body of work.

In the two decades that followed the initial Borden field studies, there was a growth in research on the underlying relationship between sedimentary architecture and the intrinsic natural variation in attributes affecting subsurface transport (e.g. Fraser and Davis, 1998; Davis

and Webb, 1998). Coordinated hydrogeologic and sedimentologic field studies created new highly-resolved data sets, at a number of field sites new and old, located in different depositional settings. This included highly resolved data characterizing the sedimentary architecture in fluvial, alluvial, and lake shoreline deposits. The studies have created a body of literature studying the link between sedimentary architecture and heterogeneity in attributes such as permeability and sorptive properties (e.g., Anderson, 1989, 1991a, 1991b; Davis et al., 1993; Davis et al., 1997; Allen-King et al., 1998; Barrash and Clemo, 2002; Divine, 2002; Guad et al., 2004; Biteman et al., 2004; Dai et al., 2005; Kalinovich et al., 2012; Ritzi et al., 2013; Soltanian and Ritzi, 2014, and Allen-King et al., 2015). In these studies, data populations were subdivided according to facies type. Facies are three-dimensional bodies of sediment whose differentiation provides a useful conceptual framework within which to characterize heterogeneity in attributes of interest within the larger sedimentary deposit. Depositional processes give rise to facies which differ in texture (grain size, sorting, and packing which affect permeability) and in mineralogy (which affects biogeochemical attributes including those controlling sorption). Importantly, facies classifications are not unique. For example, Dai et al. (2005) demonstrated how either depositional sedimentary facies or textural sedimentary facies could be used to define hydrofacies (i.e. facies usefully defining subpopulations of k) in a study of heterogeneity arising within alluvial deposits in the Rio Grande Valley. A classification should be useful and parsimonious, and these both depend upon context. What is most appropriate in one context may not be most appropriate for another. The methodologies reviewed here could be applied to any of the different facies classifications used among the studies cited above. They allow relating the geostatistics of an attribute to the specific facies classification chosen.

Two landmark contributions in the development of facies-based geostatistics were 1) the use of indicator geostatistics by Johnson and Driess (1989) to represent sedimentary architecture, and 2) the formal, combined use of continuous and indicator geostatistics by Rubin (1995) in the framework of characterizing permeability correlation. In these approaches, $I_i(x)$ is a discrete indicator space function assuming the value of 1 if x falls within facies type i , and 0 otherwise, for $i=1,2, \dots N$. An attribute such as log-permeability, $Y(x)$, is represented by the spatial function $Y(x) = \sum_i I_i(x)Y_i(x)$. Very useful spatial-bivariate sample statistics, reviewed in the next section, have since been developed.

Importantly, sedimentary architecture is hierarchical, as illustrated in Figure 1. Here we refer to a discrete structural hierarchy (*sensu* Cushman, 1990), in which larger-scale facies types (e.g. bounded by larger-scale stratification in Figure 1) comprise assemblages of smaller-scale, mutually exclusive facies types (bound by smaller-scale stratification) which, in turn, comprise still smaller-scale facies, and so on (Note that works such as di Federico and Neuman (1998) involve a different, more abstract conceptual hierarchy in which facies are not mutually exclusive and have infinite length, as clarified in Cushman's (1990) discussion. Our focus is only on discrete structural hierarchies, as used by sedimentologists to conceptualize specific preserved deposits and as illustrated in Figures 1 and 2). Statistics for spatial-bivariate correlation in discrete structural hierarchies have been formulated so that any number of hierarchical levels can be represented. These equations have been applied to highly resolved data sets used in a number of deterministic geostatistic studies at different locations (Rubin (1995), Barrash and Clemo (2002), Rubin (2003), Ritzi et al. (2004); Dai et al. (2005), Ramanathan et al. (2010) and Ritzi et al. (2007, 2013)). From these collective studies, a more fundamental understanding of the spatial-bivariate correlation for attributes controlling mass

transport has emerged and, in turn, we are gaining a more fundamental understanding of those transport processes, as reviewed below.

In the next section, the central equations used in the deterministic geostatistics are reviewed. Following that, we discuss what we believe to be the single most important insight gained from deterministic studies of spatial correlation at highly resolved field sites.

2. Useful Sample Statistics for Deterministic Studies of Spatial-bivariate Correlation

Here some relevant equations are given in a general form appropriate for both the autosemivariogram and the cross semivariogram. Consider the bounded sample domain $\mathcal{D}$, of any dimension. The symbol $\mathcal{S}$ will be used to refer to the set of sample locations within $\mathcal{D}$ at which two spatially distributed attributes ξ and ζ have been measured:

$$\mathcal{S}: \{\underline{x} | \xi(\underline{x}) \text{ and } \zeta(\underline{x}) \text{ are known and } \underline{x} \in \mathcal{D}\} \quad (1)$$

Consider a lag vector, $\underline{h}$, for defining sample lag pairs in $\mathcal{S}$ with tail at $\underline{x}$ and head at $\underline{x}'$. $N_{xx'}(\underline{h})$ is the number of sample lag pairs within $\mathcal{S}$. The general equation for the sample auto- or cross semivariogram is:

$$\hat{\gamma}_{\xi\zeta}(\underline{h}) = \frac{1}{2N_{xx'}(\underline{h})} \sum_n^{N_{xx'}(\underline{h})} (\xi(\underline{x}_n) - \xi(\underline{x}_n'))(\zeta(\underline{x}_n) - \zeta(\underline{x}_n')) \quad (2)$$

(Deutsch and Journel, 1998). For the autosemivariogram, ξ and ζ are both samples of the same attribute (e.g. log permeability). For the cross semivariogram ξ and ζ represent two different attributes (e.g. the log-permeability, Y , and the log sorption coefficient, Ξ).

Equation (2) has been expanded by including indicator spatial functions to track the facies sampled by each element of $\mathcal{S}$ and thereby track the hierarchy of facies types within the

equations (Rubin, 1995, 2003; Barrash and Clemo, 2002; Ritzi et al., 2004; Dai et al., 2005; Ritzi et al., 2013). While any number of hierarchical levels can be tracked, here we consider a domain comprising facies types defined at each of two scales (a two-level hierarchy), as illustrated in Figure 2. Consider that location $\underline{x}$ is within level I (smaller-scale) facies type o which, in turn, is within larger-scale level II facies type r (the location will be referred to as in region type ro hereafter). Let $\xi_{ro}(\underline{x}_n)$ be a sample of an attribute within region type ro and $\zeta_{ji}(\underline{x}_n)$ be a sample within region ji . Equation (2) can then be rewritten as a sample bivariate statistic designating the region types sampled by the heads and tails of $\underline{h}$:

$$\hat{\gamma}_{\xi\zeta}(\underline{h}) = \sum_r \sum_o \sum_j \sum_i \hat{\gamma}_{\xi_{ro}\zeta_{ji}}(\underline{h}) \hat{p}_{ro}(\underline{h}) \hat{t}_{ro,ji}(\underline{h}) \quad (3)$$

where the $\hat{\gamma}_{\xi_{ro}\zeta_{ji}}(\underline{h})$ are facies specific auto- or cross semivariograms (Ritzi et al., 2013). The $\hat{t}_{ro,ji}(\underline{h})$ are the sample transition probabilities which give the fraction of lags of a specific distance and direction that are of a particular transition type (i.e., the fraction with tail in region type ro and head in region type ji). The proportions $\hat{p}_{ro}(\underline{h})$ give the fraction of lags that start in region type ro .

In application the sample data are divided into subpopulations by facies type, $\xi_{ro}(\underline{x}_n)$ and $\zeta_{ji}(\underline{x}_n)$, and are used to compute the $\hat{\gamma}_{\xi_{ro}\zeta_{ji}}(\underline{h})$ terms, and co-located indicator data, $I_{ro}(\underline{x}_n)$, are used to independently compute the $\hat{p}_{ro}(\underline{h})$ and the $\hat{t}_{ro,ji}(\underline{h})$ terms of equation (3). If properly computed, the sample statistic in equation (3) should exactly match the sample statistic in equation (2) at all lags (a fact which provides for a convenient check).

The power of using this sample statistic comes from separating and analyzing different terms in equation (3), especially the transition probabilities, to the total sum and thereby assessing the relative contribution of a term and thus its significance in defining the spatial correlation. As this has been done in past studies, using a number of different data sets from different sites, some common outcomes have been observed, as described in the next section.

3. The Most Important Insight Gained from Deterministic Geostatistics at Highly Resolved Field Sites

The sample statistic in equation (3) has been computed and studied using a number of the highly-resolved data sets published in the literature. These data sets have been taken from sedimentary deposits representing a range of depositional settings including fluvial deposits (Ritzi et al., 2004), alluvial deposits (Dai et al., 2004), and lake-shore deposits (Ritzi et al., 2013). Importantly, there are some common aspects to all outcomes thus far. This group of studies is not comprehensive enough to allow sweeping generalization, but the conditions are varied enough among the different sites to suggest that the results common among these studies are outcomes that transcend the local conditions at any one of the sites, and might be common to other locations of interest as well. In all cases of application thus far, the following common results have been observed:

1) In all cases, the auto-transition terms (both $r=o$ and $i=j$) in the summation of equation (3) were exceedingly small and could be neglected.

2) In all cases the variation in the $\hat{\gamma}_{\xi\zeta_{ji}}(h)$ terms with lag was not significant to the summation at any lag and thus those terms were well approximated by constants. In other words, only the

$\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$ terms were significant to the structure (i.e. the shape and the correlation range) of the semivariogram.

3) Following from the common results (1) and (2), in all cases, equation (3) was very-well approximated by:

$$\hat{\gamma}_{\xi\zeta}(\underline{h}) \cong \sum_r \sum_j \sum_o \sum_{i \neq o} \frac{1}{2} [\hat{\sigma}_{\xi_{ro}\zeta_{ro}} + \hat{\sigma}_{\xi_{ji}\zeta_{ji}} + (\hat{m}_{\xi_{ro}} - \hat{m}_{\xi_{ji}})(\hat{m}_{\zeta_{ro}} - \hat{m}_{\zeta_{ji}})] \hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$$

(4)

where the $\hat{m}_{\xi_{ro}}$ are the univariate sample means and $\hat{\sigma}_{\xi_{ro}\zeta_{ro}}$ are the univariate (point) sample variances ($\xi = \zeta$) or point covariances ($\xi \neq \zeta$) computed from subpopulations of the data per level I facies type. In other words, the composite auto and cross sample semivariograms were very-well approximated using only the spatial-bivariate structure of the $\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$ for cross-transition probabilities. Thus, the semivariograms grew with lag as grew the sample probability of transitioning across different facies types, and stabilized with lag as did those sample cross-transition probabilities.

The significance of this third result is illustrated in Figure 3 using the set of results from the Borden research site (Ritzi et al., 2013), where the data included co-located measurements of log permeability, Y , the log sorption distribution coefficient for PCE, Ξ , and integer indicator data for the sampled facies type at each of two hierarchical levels (i.e., the facies types illustrated in Figure 2). Details of the data sets are given in (Ritzi et al., 2013 and Allen-King et al., 2015). Figure 3 shows the comparison of the approximation in equation 4 (dashed line) to the composite sample semivariogram given both by equation 2 (solid line) and equivalently by equation 3 (circles). The comparisons for the sample semivariogram of log permeability $\hat{\gamma}_{YY}(\underline{h})$, for the log

distribution coefficient $\hat{\gamma}_{\Xi\Xi}(\underline{h})$, and for the $\hat{\gamma}_{\Xi Y}(\underline{h})$ cross semivariogram are all quite close, indicating that the approximation in equation (4) represents the sample correlation structure (i.e, the shape of the rise and the correlation range) quite well. Importantly, equation (4) was computed with the identical set of $\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$ cross-transition terms for all three cases: $\hat{\gamma}_{YY}(\underline{h})$, $\hat{\gamma}_{\Xi\Xi}(\underline{h})$, and $\hat{\gamma}_{\Xi Y}(\underline{h})$. Therefore, the underlying shape of the spatial-bivariate structure of all three sample semivariograms is defined by the common sedimentary architecture, and furthermore, this spatial-bivariate correlation structure can be inferred from the $I_{ro}(\underline{x}_n)$ data (relatively independently from spatially distributed data for Y and Ξ , also see Figures 4a&b in Ritzi et al., 2013). Importantly, note that the structure common to $\hat{\gamma}_{YY}(\underline{h})$, $\hat{\gamma}_{\Xi\Xi}(\underline{h})$, and $\hat{\gamma}_{\Xi Y}(\underline{h})$ is not arising from uniform point correlation between Y and Ξ , assumed in many prior articles. As explained by Kalinovich et al. (2012), Ξ is related to abundance of dark carbonate lithocomponents containing kerogen within the mineral matrix, and $Y - \Xi$ point correlation is not uniform and varies from weak positive to weak negative among the facies types (Ritzi et al., 2013; Allen-King et al., 2015). Again, the structure common to $\hat{\gamma}_{YY}(\underline{h})$, $\hat{\gamma}_{\Xi\Xi}(\underline{h})$, and $\hat{\gamma}_{\Xi Y}(\underline{h})$ arises from the common sedimentary architecture.

Thus, even at the mildly heterogeneous Borden research site, the sedimentary architecture (here of a lake-shore deposit) is fundamental to a basic understanding of the spatial correlation underlying the attributes controlling mass transport processes. The cross-transition probability structure has also been shown to define the permeability semivariogram structure in this way in more heterogeneous alluvial deposits (e.g. Dai et al., 2005, their Figures 5 and 6), and in fluvial deposits (e.g. Ritzi et al., 2004, their Figure 10). Thus, there is a growing body of work suggesting that it is the cross-transition probability structure, defined by hierarchical stratal

architecture, that commonly (though perhaps not exclusively) is the foundation for understanding the spatial correlation of attributes controlling mass transport processes.

4. Discussion: Benefits from this Insight, both to Basic Science Understanding and to Practical Application

Separating the influence of each scale of sedimentary architecture, and identifying the relative contribution of each scale to a transport process, provides a scientific foundation upon which the basic-science understanding of a transport process is improved. Furthermore, the insights gained at highly-resolved, data-rich research sites provide practical benefits to addressing the common challenges created by data limitations in engineering application by practitioners at specific environmental cleanup sites. Here both the benefit to basic science understanding and the benefit to practitioners are reviewed and discussed.

Given that the $\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$ cross-transition probability structure defines the structure of spatial-bivariate correlation for hydraulic/reactive attributes like Y and Ξ , as demonstrated for the Borden site above, it is possible to separate and study the influence of different scales of sedimentary architecture. For example, carrying forward with the example above, transitions across the different smaller-scale facies types (i.e. transitions from region type ro to region type ri , $\forall o \neq i$) can be separated from transitions across the different larger-scale facies types (i.e. transitions from region type ro to region type ji , $\forall r \neq j, o \neq i$), thus isolating the correlation arising from the different scales of sedimentary architecture. Furthermore, analytical Lagrangian-based transport models (e.g. Dagan, 1991) are generally linear in their underlying correlation models. Therefore, decomposing the correlation model also allows decomposing the transport

models, facilitating the study of the relative contributions of different scales of sedimentary architecture to a mass transport process. This decomposition can improve the fundamental understanding of that transport process.

The summation terms in equation (4) can be decomposed and organized in many different ways. One way, proven to be useful among all of these studies, is to simply separate them by hierarchical level. Results from the Borden research site are briefly reviewed here for illustration. The hierarchy of facies types defined for the Borden aquifer gives, at any single non-zero lag, 18 possible types of transitions across level I facies types ($o \neq i$) and 24 possible types of transitions across level II facies types ($r \neq j, o \neq i$). We can separate these groups of summation terms to isolate the relative influence of each scale (level I vs. level II) as follows:

$$\begin{aligned} \hat{\gamma}_{\xi\xi'}(\underline{h}) &\equiv (A_{\xi\xi'} + B_{\xi\xi'})\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ri}(\underline{h}) \\ &+ (C_{\xi\xi'} + D_{\xi\xi'})\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h}) \end{aligned} \quad (5)$$

where

$$A_{\xi\xi'} = \sum_r \sum_o \sum_{i \neq o} \frac{1}{2} [\hat{\sigma}_{\xi_{ro}\xi_{ro}} + \hat{\sigma}_{\xi_{ri}\xi_{ri}}] \hat{p}_{ro} \hat{p}_{ri} ,$$

$$B_{\xi\xi'} = \sum_r \sum_o \sum_{i \neq o} \frac{1}{2} [(\hat{m}_{\xi_{ro}} - \hat{m}_{\xi_{ri}})(\hat{m}_{\xi_{ro}} - \hat{m}_{\xi_{ri}})] \hat{p}_{ro} \hat{p}_{ri} ,$$

$$C_{\xi\xi'} = \sum_r \sum_{j \neq r} \sum_o \sum_{i \neq o} \frac{1}{2} [\hat{\sigma}_{\xi_{ro}\xi_{ro}} + \hat{\sigma}_{\xi_{ji}\xi_{ji}}] \hat{p}_{ro} \hat{p}_{ji} ,$$

$$D_{\xi\xi'} = \sum_r \sum_{j \neq r} \sum_o \sum_{i \neq o} \frac{1}{2} [(\hat{m}_{\xi_{ro}} - \hat{m}_{\xi_{ji}})(\hat{m}_{\xi_{ro}} - \hat{m}_{\xi_{ji}})] \hat{p}_{ro} \hat{p}_{ji} ,$$

The first of the two groups on the RHS of equation (5) is referred to as the $\alpha\chi$ cross-transition group, indicating that its terms are for lags that are auto-transitions (α) at level II and cross-transitions (χ) at level I. That is to say, its terms are only for lag transitions across different level I facies types occurring within the same level II facies type. The second group on the RHS of equation (5) is referred to as the $\chi\chi$ cross-transition group, indicating that its terms are for lags that are transitions across facies types which are different at both level I *and* level II. Isolating the $\alpha\chi$ and $\chi\chi$ groups allows for separating and studying the influence of each scale of sedimentary architecture. We expect, of course, that the correlation range for $\chi\chi$ cross-transitions will be larger than for $\alpha\chi$ cross-transitions. Furthermore, we can separate the influence of the difference in mean value at the tail and head of these transitions, from the influence of variability around the mean value at the tail or head. At the tail and head of $\alpha\chi$ cross-transitions, term $B_{\xi\xi'}$ represents the influence imparted by the difference in the means, and term $A_{\xi\xi'}$ represents the influence of the point (co)variation about the mean within the level I facies at each end. At the tail and head of $\chi\chi$ cross-transitions, term $D_{\xi\xi'}$ represents the influence imparted by the difference in the means, and term $C_{\xi\xi'}$ represents the influence of the point (co)variation about the mean within larger level II facies at each end.

The shape of $\hat{p}_{ro}(\underline{h})\hat{t}_{ro,ji}(\underline{h})$ cross-transition probabilities is known to be strongly determined by the variance in the length of facies types (Ritzi, 2000). Facies length distributions, when fully characterized without sample bias, are typically Erlangian with a coefficient of variation of the order of unity, which gives rise to asymptotic and exponential-like transition probability structures (White and Willis, 2000; Ritzi, 2000). Accordingly, in the various field studies cited above, including at the Borden site, the sample cross-transition probability

structures indeed had asymptotic exponential-like shapes. Based strictly on the quantified coefficient in variation in sample length, not fitting, exponential structures were chosen as models, parsimoniously using one exponential structure for each level of the hierarchy. The effective ranges at each hierarchy level are related to facies proportions and lengths at each level by:

$$a_{\alpha\chi} = \sum_o \sum_r 3\bar{l}_{ro}(1 - \hat{p}_{ro}) \hat{p}_{ro} \quad (6a)$$

and

$$a_{\chi\chi} = \sum_r 3\bar{l}_r(1 - \hat{p}_r) \hat{p}_r \quad (6b)$$

where $\bar{l}_{ro}$ and $\bar{l}_r$ are the mean length of facies types defined at level I and level II respectively (Carle and Fogg, 1996; Carle, 1996; Ritzi, 2000).

For the Borden example being used here to illustrate these points, univariate statistics for proportions and length of facies types at level I and level II were given by Ramanathan et al. (2010). The stratal architecture, and these statistics governed by it, give rise to a smaller effective correlation range equal to 3.6 m for $\alpha\chi$ cross-transition probabilities, and a larger range of 6.3 m for $\chi\chi$ cross-transition probabilities. Univariate statistics for Y and Ξ are given in Ritzi et al. (2013; Table 2), and are not repeated here. When the effective correlation ranges and A - D terms are computed from these univariate statistics, semivariogram models match the sample semivariograms quite well, as summarized in Figure 4. Furthermore, Ramathan et al. (2010) derived a Lagrangian-based model for the dispersion of non-reactive solutes using the correlation model for $\hat{\gamma}_{YY}(\underline{h})$. Similarly, Soltanian et al. (2015, two-part companion articles) derived models for the time-dependant retardation and dispersion for the reactive transport of PCE. These results are also summarized in Figure 4. Note that the Lagrangian transport models

represent average behavior among the ensemble of possible realizations from the correlation model, and thus we don't expect an exact match to the single realization captured in the Borden field experiment, only a representation of the general behavior (see expanded discussion in the cited works). As illustrated in Figure 4, in all cases, the semivariogram models and the Lagrangian-based transport models do represent the general behavior quite well. Note this is accomplished without using any fitting parameters; all model parameters are physically quantifiable attributes, directly and independently measured from the sediment cores.

By having decomposed the spatial correlation structure in this way, the relative contribution from each scale of sedimentary architecture to defining the shape and range of the spatial correlation structure is clear (Figure 4, left three panels). Accordingly, the separate contribution from each scale to defining the magnitude and duration of transport processes is also clear (Figure 4, right three panels). The larger-scale facies types defined at level II create a longer correlation range in the semivariograms than is created by the smaller-scale level-I facies types, as seen by examining the larger effective range of the $\chi\chi$ curves (Figure 4, left three panels). Furthermore, the larger difference in the mean values across level II facies types, and the larger variation around the mean, are the reasons that there is a larger contribution to the magnitude of the semivariograms from the level II facies types relative to the contribution from level I types, as clearly shown by the larger magnitude of the $\chi\chi$ curves. Accordingly, the correlation range of the level II architecture controls the ultimate *duration* of the time-dependent changes in the effective retardation factor for the PCE plume, and the duration of the time-dependent changes in the rate of plume spreading of all the plumes. The duration of time-dependency in the PCE retardation factor is the most visible of these and, as shown by Soltanian et al. (2015, Part 1), is exactly the time needed for the plume centroid to travel the distance equal

to the correlation range of the larger-scale, level II cross-transition probability structure. The duration of the time-changing rate of spreading of the plumes is also determined by the level II range, and because the rate of spreading relates to velocity variation rather than average velocity, it changes until all particles (not just the average particle) have fully sampled the level-II heterogeneity. The *magnitude* of the level II scale contribution to the semivariogram at any lag is larger than the level I scale contribution, and thus level II contributes more to the *magnitude* of the time-dependent retardation and dispersion. This is shown in Figure 4 by the significantly greater relative contribution of the $\chi\chi$ curves in defining the models. The works cited above have further decomposed the models in order to study the relative contributions of the *A-D* terms in the above model, showing that the magnitude of these transport processes can be mostly explained by differences in means across the larger-scale level-II facies types. Further analysis is provided in Soltanian et al. (2015, two-part companion articles).

Importantly, the newer studies and newer models do not necessarily lead to more accurate predictions of the transport behavior. The main benefit derived from the newer approach is that it leads to a more fundamental understanding of the transport processes. Models developed in earlier studies represented tracer plume behavior quite well, but the correlation range was determined by curve fitting of the sample bivariate statistic. Thus, the fundamental controlling parameter was a fitting parameter. There was speculation about relationships to sedimentary architecture, but no fundamental understanding in the original Borden site studies (Sudicky, 1986; Woodbury and Sudicky, 1991, see also conjectures in discussion by Dagan, 1989a,b; Naff et al., 1988, 1989; and Rajaram and Gelhar, 1991). The newer studies that characterized, quantified and directly represented the hierarchical sedimentary architecture in the equations showed that the transport processes can be explained mostly by the occurrence of larger-scale

facies comprising medium sand, juxtaposed with larger-scale facies comprising finer-grained material, mostly fine sand. Because the transport processes are understood in a more fundamental way, the newer models can be parameterized without curve fitting.

In addition to the more fundamental understanding gained through this approach, there is perhaps knowledge that would be of value to practitioners. One potential benefit is that the approach provides a rational basis for explaining and modeling the spatial correlation of the attributes. Modeling the spatial-bivariate correlation can be problematic even at data rich sites. As elucidated by Woodbury and Sudicky (1991), even with large amounts of permeability data it is “unlikely that one can unequivocally distinguish between various competing variogram models” by curve fitting. However, in the newer approach, by using the indicator space functions to represent stratal architecture and representing correlation through the cross-transition probability structures, the variogram model is based simply on statistics for proportions and lengths of facies types. Thus, all model parameters are independent and physically quantifiable attributes of the stratal architecture. Correlation can be modeled without fitting sample bivariate statistics for either indicator or continuous space variables. There is a rational basis for representing the parameter covariance, which gets around the equivocal aspects of model fitting raised by Woodbury and Sudicky (1991). Another potential practical benefit comes from the fact that commonly there are challenges created by data limitations at specific environmental cleanup sites. Because of time, effort and cost, K_d data are the most difficult to acquire and the least likely to be available, followed by permeability data. On the other hand, geologic information can sometimes be directly observed in drill cuttings or cores, and facies proportions and lengths can be quantified from outcrop exposures, cores, geophysical transects, or inferred from stratigraphic models, as in examples by Dai et al. (2005), Lunt et al. (2005),

Rubin et al. (2006) and Ritzi and Allen-King (2007). As a result, geologic data are typically more abundant than permeability or K_d data. Therefore there is strong practical benefit in being able to directly use geologic data in evaluating transport processes at sites with limited data. The newer approach requires less of the harder-earned permeability and K_d data. Note that Sudicky's (1986) seminal analyses of solute spreading at the Borden site involved determining sample bivariate statistics for permeability with use of 1279 data, almost an order of magnitude more in number than the number of permeability data used by Ritzi et al. (2013). Because the correlation model in the newer approach is based on geologic indicator data, and therefore only univariate statistics for permeability are required, the permeability data requirements were significantly reduced in the latter study.

However, the approach does not by any means fully solve the general problem of data limitations. For example, as discussed by Dai et al. (2005), outcrop exposures of aquifer sediment, while useful for characterizing sedimentary architecture and quantifying important aspects of it, are not a panacea. Often these exposures are of limited extent and the resulting bias in facies lengths must be accounted for. Bias in length imparted by incomplete exposures can be reduced in some cases by accounting for transition frequencies (White and Willis, 2000; Dai et al., 2005). A corollary is that permeability semivariograms, if determined traditionally, directly from spatially distributed permeability data, will not be representative unless the sample locations also allow an adequate determination of the cross-transition probability structure (e.g. Popken (2005) examined this at the Oyster research site).

Summary and Conclusions

Naturally-occurring sedimentary deposits commonly exhibit multiscaled and hierarchical stratification. That is, stratification can be delineated at different scales, and units delineated at one scale comprise assemblages of units delineated at the next smaller scale, and so on, in a discrete structural hierarchy. The spatial-bivariate correlation of permeability in such deposits can be directly and quantitatively linked to the multiscaled and hierarchical stratification. The same can be true for some other variables affecting subsurface transport. Research on this topic has been facilitated by highly-resolved data sets developed at different research sites located in a variety of depositional environments (lake-shore, alluvial, fluvial).

At all of these research sites, the sample semivariogram:

$$\hat{\gamma}_{\xi\xi'}(\underline{h}) = \frac{1}{2N_{\xi\xi'}(\underline{h})} \sum_n^{N_{\xi\xi'}(\underline{h})} (\xi(\underline{x}_n) - \xi(\underline{x}_n'))(\xi(\underline{x}_n) - \xi(\underline{x}_n')) = \sum_o \sum_i \hat{\gamma}_{\xi_o\xi_i}(\underline{h}) \hat{p}_o(\underline{h}) \hat{t}_{o,i}(\underline{h})$$

was very-closely matched by the approximation:

$$\hat{\gamma}_{\xi\xi'}(\underline{h}) \cong \sum_o \sum_{i \neq o} \frac{1}{2} [\hat{\sigma}_{\xi_o\xi_o} + \hat{\sigma}_{\xi_i\xi_i} + (\hat{m}_{\xi_o} - \hat{m}_{\xi_i})(\hat{m}_{\xi_o} - \hat{m}_{\xi_i})] \hat{p}_o(\underline{h}) \hat{t}_{o,i}(\underline{h})$$

demonstrating that the sample semivariogram structure (i.e. shape and correlation range) was almost entirely defined by the sample cross-transition probability structure, $\hat{p}_o(\underline{h}) \hat{t}_{o,i}(\underline{h})$. Thus, the structure in the spatial-bivariate correlation for permeability (i.e., the shape and range) was essentially that of the cross-transition probability structure. Cross-transition probability structures are determined by the spatial arrangement of facies within the sedimentary architecture. The shape of a cross-transition probability structure is determined by the variance in facies lengths, and the range is determined by the proportions and mean lengths of facies. Thus, at these sites, the structure in the spatial correlation for permeability (and some other attributes) could be directly and quantitatively linked to the proportions and length statistics of the facies delineated within the sedimentary architecture. Accordingly, the sample spatial-

bivariate correlation structure for permeability could be modeled using independently-quantified, univariate, sample statistics for proportions and lengths, in all of these studies.

Importantly, the direct and quantitative link between the spatial correlation structure for permeability and the cross-transition probability structure of the sedimentary architecture identified in these studies was a fundamental link made through the examination of highly-resolved sample statistics, independent of probability theory, modeling or curve fitting. The observed link might not be universal, but the conditions are varied enough among these different studies to suggest that this link may be commonly observed.

Knowing that the spatial-bivariate correlation structure of permeability (and some other hydraulic/reactive attributes) is essentially the cross-transition probability structure for the sedimentary architecture improves our basic scientific understanding of the spatial correlation for hydraulic and reactive attributes that affect subsurface transport processes and, in turn, improves our fundamental understanding of the transport processes themselves. For example, based on this knowledge it is possible to decompose the correlation structure according to the contributions from each scale. The separate influence of each scale of sedimentary architecture on the spatial correlation structure of an attribute can be studied and, therefore, the influence of each scale on a transport process can be separately identified and understood at a fundamental level. Furthermore, the influence from the variance of hydraulic/reactive attributes within facies at each scale and the contrast in the mean across facies boundaries can also be separated, studied, and their relative contributions to a transport process understood. The knowledge that the spatial-bivariate correlation structure of permeability (and some other hydraulic/reactive attributes) is essentially the cross-transition probability structure also provides possible benefits to practitioners challenged by data limitations at environmental cleanup sites. This knowledge

492 provides a formal way to directly use geologic data (more easily acquired than permeability or
493 K_d data) in estimating correlation structure and in evaluating transport processes. Correlation can
494 be modeled from independently-quantified univariate statistics, and data requirements may be
495 reduced. In conclusion, the basic scientific understanding of how correlation is defined by
496 sedimentary architecture has provides a rational basis for understanding, explaining, and
497 modeling the spatial-bivariate correlation structure of hydraulic/reactive attributes that affect
498 subsurface transport processes.

499

Figure captions:

Figure 1. An illustration of hierarchical sedimentary architecture common to fluvial deposits (modified from Lunt and Bridge, 2004). The figure shows three scales of stratification in a hierarchy in which any one scale comprises assemblages of strata types at the next lower level. In the example shown, the largest-scale of stratification bounds compound-bar deposits. These comprise medium-scale stratification bounding unit bar deposits. Unit bar deposits comprise sets of cross-strata defined by three different grain-size categories. The levels in hierarchy of stratification are numbered, arbitrarily from the smaller to the larger scale. The lower photo shows an exposure of the level I facies (from Lunt, 2002).

Figure 2. Textural facies defined at two scales, observed in sediment cores taken at the Borden site (from Ramanathan et al., 2010). Note, this site represents a different depositional environment than shown in Figure 1 and consequently has a different stratification. a) Map of the larger-scale level II unit types, and b) map of the smaller-scale level I unit types comprised by them. Maps share the same ordinate and abscissa labels. At level II, the facies are medium sand (M) and fine sand and silt (FZ). Facies M comprises, at level I, medium sand facies with distinct lamination (MLD), faint lamination (MLF), and massive with no lamination (MM). The FZ facies comprises fine sand with distinct lamination (FLD), faint lamination (FLF), no lamination (MM), and silt (Z). The example lag vector transitions at level II from facies M to facies FZ, and at level I from facies MM to facies FLF.

Figure 3. Comparison of the approximation in equation 4 (dashed line) to the composite semivariogram in equation 2 (line with circles). The comparisons for the semivariogram of log-permeability (Y), log distribution coefficient (Ξ), and the Y - Ξ cross semivariogram are all quite close, indicating that the approximation explains the correlation structure (i.e, the shape of the rise, the range, and the sill) quite well. Note that in all three cases, the structure of the approximation is the structure of the probability of transitioning across different facies types, as defined by the sedimentary architecture. The same, common, cross-transition probability structure defines the structure of all three semivariograms. Thus, even at the mildly heterogeneous Borden research site, defining the sedimentary architecture (here of a lake-shore deposit) is fundamental to attaining a basic understanding of spatial correlation structure. The cross-transition probability structure has been shown to define the semivariogram structure in this way in all other studies of sedimentary deposits in which this analysis has been applied to date, including in alluvial fan deposits (Dai et al., 2005) and in fluvial deposits (Ritzi et al., 2004).

Figure 4. Comparing models to observations. Note that none of the models are fitted. All are computed from the same, independently determined, univariate sample statistics including the proportions and mean length of facies types (defining the cross-transition probability structure, and corresponding correlation ranges at each scale), and univariate statistics for Y and Ξ (which scale the ordinate axes). Left three panels show semivariogram models (lines) compared to observations (red circles) [taken from Ritzi et al. 2013]. Right three panels show Lagrangian-based models (lines) for dispersion and retardation compared to observations (symbols including

circles and crosses) computed by Ramanathan et al. (2010) and Soltanian et al. (2-part articles, 2015).

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FIGURES

FIGURE 1

Figure 2

FIGURE 3

Figure 4

— Composite Model
— Model $\chi\chi$ Terms
— Model $\alpha\chi$ Terms