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Dispersion of groundwater age in an alluvial aquifer system

Gary S. Weissmann,[1] Y. Zhang, Eric M. LaBolle, and Graham E. Fogg[2]

Hydrologic Sciences, University of California, Davis, California, USA

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[1] Interpretation of groundwater ages typically rests on assumptions of minimal mixing of different water ages in the water samples. The effects of three-dimensional, geologic heterogeneity on groundwater mixing and tracer concentrations, however, have not been evaluated. In this study, we use a series of 10 detailed geostatistical realizations along with high-resolution numerical groundwater flow and contaminant transport simulation to model distributions of groundwater ages and chlorofluorocarbon (CFC) ages at wells within a heterogeneous stream-dominated alluvial fan aquifer system. Results show that groundwater reaching a well in the heterogeneous aquifer system typically consists of a wide distribution of groundwater ages (often spanning >50 years), even over short (<1.5 m) screened intervals. Additionally, simulated arithmetic mean groundwater ages do not correspond to mean ages estimated from simulated CFC concentrations. Results emphasize the potential ambiguity of "mean" groundwater ages estimated from environmental tracer concentrations in typically heterogeneous geologic systems. The significant dispersion of groundwater ages also implies that ultimate, maximum effects of nonpoint source, anthropogenic contamination of groundwater may not be reached until after many decades or centuries of gradual decline in groundwater quality. *INDEX TERMS:* 1829 Hydrology: Groundwater hydrology; 1832 Hydrology: Groundwater transport; *KEYWORDS:* heterogeneity, groundwater age, environmental tracer, chlorofluorocarbons, transport, modeling

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1. Introduction

[2] The use of environmental tracers, such as chlorofluorocarbons (CFCs), tritium/helium-3, and carbon-14, for age dating groundwater has made it possible to estimate mean ages of groundwater pumped from wells [e.g., *Solomon and Sudicky*, 1991; *Solomon et al.*, 1992; *Hackley et al.*, 1992; *Plummer et al.*, 1993; *Busenberg et al.*, 1993; *Cook et al.*, 1995; *Johnston et al.*, 1998; *Varni and Carrera*, 1998; *Plummer and Sprinkle*, 2001]. In concept, groundwater age dates provide important information on sources and timing of recharge and vulnerability of aquifers to contamination. An age date of 40 years, for example, might suggest that surface contaminant sources that have only existed for 20 years could not be present in the 40-year-old water. Accordingly, there is growing interest among regulatory agencies in using groundwater age dates to map susceptibility of groundwater systems to contamination.

[3] Many papers state or imply that the validity of the age-dates is suspect if dispersion and/or long screened intervals create significant mixing of ages [*Walker and Cook*, 1991; *Mazor and Nativ*, 1992; *Goode*, 1996; *Varni and Carrera*, 1998; *Tompson et al.*, 1999; *Bethke and Johnson*, 2002]. *Hinkle and Snyder* [1997] noted that mixing a small portion of younger water that contains CFC with older water that is CFC-free will result in reporting of a CFC age date that is between 1944 and the date of recharge of the modern water in the mixture, even if the water sample consists of a low percentage of younger water. Thus groundwater ages determined from CFC data may not represent the weighted mean age of groundwater where mixing is significant. *Varni and Carrera* [1998] presented similar results indicating this influence of mixing to be true for any environmental tracer in a heterogeneous aquifer. Though several writers have indicated that the influence of heterogeneity and groundwater mixing due to hydrodynamic dispersion might produce uncertainty in environmental tracer-based age date results [*Busenberg and Plummer*, 1992; *Dunkle et al.*, 1993; *Reilly et al.*, 1994; *Johnston et al.*, 1998; *Varni and Carrera*, 1998; *Burow et al.*, 1999; *Tompson et al.*, 1999], the magnitude of this effect is largely unknown. It is often assumed, however, that age dating of waters from short screened intervals (~1 m) will eliminate significant mixing of different groundwater ages from different depths.

[4] *Bethke and Johnson* [2002] used the age-mass equation of *Goode* [1996] to analytically model effects of mass transfer between regional aquifer and aquitard layers. They showed that for very long times of transport, a conservative environmental tracer that undergoes advection, dispersion, and diffusion (Brownian motion) will, in the limit, become distributed among the aquitard and

[1]Now at Department of Geological Sciences, Michigan State University, East Lansing, Michigan, USA.

[2]Also at Department of Geology, University of California, Davis, California, USA.

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aquifer materials in direct proportion to the relative volumes of those materials. Because diffusive mass transfer into the aquitards must be balanced by diffusive mass transfer out of the aquitards, mean groundwater ages in the aquifers can be substantially older than ages that would be indicated by a simple piston displacement model for the aquifer alone.

[5] The purpose of our work is to examine the role of dispersion on groundwater age dates in an alluvial aquifer complex containing typical heterogeneity in hydraulic conductivity. While *Bethke and Johnson* [2002] examined steady state effects of age mixing among aquifer and aquitard layers, wherein diffusion is the dominant transport process in the aquitards, we examine the effects of advection, dispersion, and diffusion in a multiscale model of heterogeneity containing multiple hydrostratigraphic units, some of which are dominated by diffusion. *Bethke and Johnson*'s [2002] analysis is most applicable for understanding potential age mixing of old groundwaters (say, >10,000 yr) dated by radiometric tracers that have reached a steady state in terms of mass transfer between tight aquitards and the aquifers. In contrast, the present study applies more to dating of younger groundwaters with environmental tracers that have not migrated long enough or far enough to have reached equilibrium in terms of mass exchange among the various hydrostratigraphic units.

[6] We also explore whether, as often suggested in the age-date literature, the age distribution is appropriately narrow when the screened interval is small. The literature contains limited quantitative evaluations of these assumptions with field data or with models [e.g., *Varni and Carrera*, 1998; *Tompson et al.*, 1999]. In particular, effects of dispersion due to heterogeneity and mixing due to well screen length on age dates has, to our knowledge, never been investigated in typically complex alluvial materials.

2. Study Area

[7] We tested the effects of heterogeneity on groundwater age distributions in the Kings River Alluvial Fan aquifer system, located southeast of Fresno, California (Figure 1). The Kings River deposited a stream-dominated alluvial fan where it exits the Sierra Nevada into the San Joaquin Valley, thus the aquifer consists of a highly heterogeneous mix of lithologies ranging from cobble to mud (i.e., silt and clay) [*Weissmann*, 1999; *Weissmann and Fogg*, 1999; *Weissmann et al.*, 2002]. Five hydrofacies were recognized within this aquifer system based on core descriptions. These include, in decreasing order of hydraulic conductivity, gravel, sand, muddy sand, mud, and paleosol [*Burow et al.*, 1997; *Weissmann*, 1999; *Weissmann and Fogg*, 1999]. Additionally, stratigraphic studies show that the aquifer consists of five depositional sequences, where each sequence consists of a heterogeneous composite of the gravel, sand, muddy sand, and mud hydrofacies, and each sequence is bounded by laterally continuous unconformities marked by the paleosol hydrofacies [*Weissmann*, 1999; *Weissmann and Fogg*, 1999; *Weissmann et al.*, 2002].

[8] *Burow et al.* [1999] reported collection of CFC samples in 1994 and 1995 from several discrete intervals within the Kings River alluvial fan. Twenty multilevel nested sampling wells at six locations (B1, B2, B2.5, B3, B4, and B5) were used for this sampling. CFC-based age dates indicate that groundwater ages range between 2 and >55 years at the time of this 1994-95 sampling (Table 1).

3. CFC Age Dating Method

[9] Age dating of groundwater with CFC is based on correlation of variable atmospheric tracer concentrations through time to tracer concentrations in the groundwater at sampling locations. For instance, CFC-11 (CCl_3F) and CFC-12 (CCL_2F_{-2}) concentrations in the atmosphere have steadily increased since the 1940s (Figure 2) [*Elkins et al.*, 1993, 1999; *Plummer et al.*, 1993]. CFCs are soluble in water, and the CFC concentration in water (C_{CFC}) is related to the atmospheric CFC partial pressure by Henry's Law

$$C_{CFC} = K_{CFC,T} P_{CFC} \tag{1}$$

where $K_{CFC,T}$ is the Henry's Law constant for the CFC compound at temperature, T, and P_{CFC} is the atmospheric partial pressure of the CFC compound [*Warner and Weiss*, 1985; *Plummer et al.*, 1993]. Assuming meteoric water recharged to the groundwater is at equilibrium with the atmosphere with respect to the CFCs, estimates of CFC partial pressure in the atmosphere at time of recharge can be obtained from groundwater sample CFC concentrations by application of equation (1). A groundwater age is estimated by relating this estimated past CFC partial pressure to historic CFC atmospheric concentrations shown in Figure 2 [*Plummer et al.*, 1993; *Busenberg et al.*, 1993].

[10] Several assumptions in estimating groundwater ages from CFC data are outlined by *Plummer et al.* [1993], including (1) the recharge temperature is known, (2) collection procedures do not allow contact with the atmosphere or other sources of CFCs, (3) no local sources of CFC contamination in the groundwater exist outside of the background atmospheric levels, and (4) groundwater concentrations are not significantly affected by hydrodynamic dispersion, thus piston-type transport of the tracer can be used to approximate tracer movement in the groundwater system.

4. Modeling Heterogeneity, Groundwater Flow, and Environmental Tracer Transport

[11] The modeling approach used to understand the influence of heterogeneity on groundwater ages and CFC concentrations at a well was accomplished in three steps: (1) model the heterogeneity through application of transition probability geostatistics within a sequence stratigraphic framework [*Weissmann and Fogg*, 1999], (2) simulate steady state groundwater flow using a block-centered finite difference model, and (3) estimate water travel time from the water table to wells using backward-in-time random walk particle tracking [*Uffink*, 1989; *Fogg et al.*, 1999; *LaBolle et al.*, 2000]. The following sections outline this approach.

4.1. Modeling Heterogeneity

[12] To simulate the heterogeneous distribution of hydrofacies within the Kings River alluvial fan aquifer system, we used a transition probability geostatistics approach [*Carle and Fogg*, 1996; *Carle et al.*, 1998; *Weissmann et al.*, 1999]. *Weissmann and Fogg* [1999] describe application

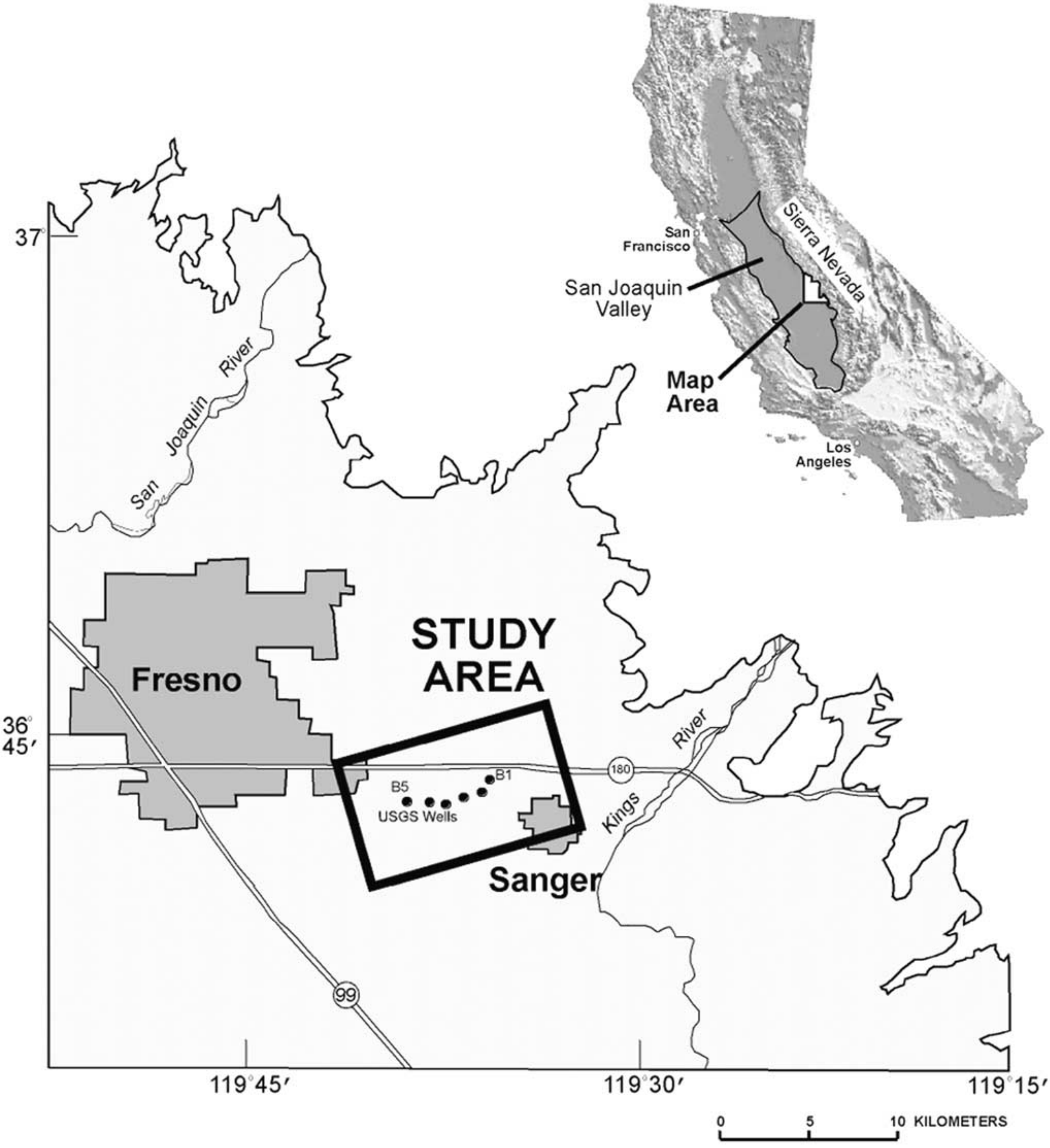

Figure 1. A map showing the study area location southeast of Fresno, California. The six USGS well nest locations are indicated in the study area boundaries (modified from *Burow et al.* [1999]).

of this approach to the Kings River Alluvial Fan study area. The following discussion is a brief summary of the geostatistical work.

[13] In the transition probability geostatistical approach, transition probabilities between categorical data (e.g., hydrofacies) are measured in the vertical and horizontal directions and a Markov chain model is fit to these measured results. The 3-D Markov chain model is then used in sequential indicator simulation followed by simulated annealing to produce realizations of the subsurface heterogeneity. Importantly, "soft" or interpreted geologic information, such as facies mean lengths, global facies proportions, and facies juxtaposition relationships, can be incorporated into the Markov chain model of spatial variability [*Carle et al.*, 1998]. Because close-spaced data are plentiful in the vertical direction from core and geophysical well logs, fitting a model to measured vertical transition probabilities is straightforward. To estimate the more difficult horizontal components of the Markov chain model, we analyzed C-horizon textures from soil surveys in a geologic

Table 1. Reported CFC Age Dates From USGS Wells Within the Study Area Compared to Mean Simulated CFC Age Dates for the 10 Realizations From This Study[a]

Well and Screen Number	Screen Midpoint Elevation, m amsl	Simulated Residence Time, years		Reported CFC Ages, years		Simulated CFC Ages, years			
		Mean	SD	CFC-11	CFC-12	CFC-11		CFC-12	
						Mean	SD	Mean	SD
B1-1	85.2	22.4	11.3	20	8	20.6	8.7	20.7	9.1
B1-2	58.7	a	a	38	41	a	a	a	a
B1-3	28.2	a	a	37	48	a	a	a	a
B2-1	84.3	31.6	14.7	16	2	14.0	3.7	13.7	3.8
B2-2	81.8	33.3	16.1	8	4	14.4	3.9	14.1	3.9
B2-3	68.1	57.2	12.5	22	18	26.7	4.5	27.8	4.8
B2.5-1	64.2	81.0	32.2	31	39	33.6	12.0	34.9	12.4
B2.5-2	52.9	144.6	64.8	29	32	49.9	6.0	51.1	4.6
B3-1	85.2	17.7	9.6	6	3	9.7	2.2	9.4	2.3
B3-2	72.1	34.2	11.7	b	16	17.3	3.1	17.5	2.9
B3-3	54.1	87.0	37.7	32	33	32.6	5.5	34.4	6.4
B3-4	46.5	96.9	38.9	29	41	38.1	7.8	40.5	8.9
B3-5	25.8	a	a	35	>55	a	a	a	a
B4-1	81.2	18.8	9.2	7	2	10.9	3.3	10.7	3.6
B4-2	70.0	43.6	16.3	b	b	18.7	2.2	18.9	2.5
B4-3	48.9	104.4	33.1	b	b	36.4	3.9	39.5	4.7
B4-4	25.4	124.7	39.0	b	30	45.3	4.0	49.3	4.0
B5-1	78.2	23.9	18.3	14	6	14.3	7.7	14.5	8.0
B5-2	54.4	52.1	18.1	26	25	25.4	4.9	26.4	5.5
B5-3	20.9	124.5	32.4	b	b	49.7	4.8	52.2	3.1

[a]Also included are simulated mean ages of groundwater at selected wells. Reported CFC ages are from *Burow et al.* [1999]. Here a indicates that significant particles through upgradient boundary (>15%), and therefore groundwater age was indeterminate; b indicates that measured CFC-based age dates were not reported by *Burow et al.* [1999].

depositional systems context, as described in *Weissmann et al.* [1999] and *Weissmann and Fogg* [1999].

[14] To deal with the nonstationarity created by the multiple stratigraphic sequences and to represent discontinuity across the unconformity boundaries, we modeled each stratigraphic sequence separately [*Weissmann and Fogg*, 1999]. Additionally, the paleosol hydrofacies marks the laterally continuous unconformity surface and was formed over the exposed fan surface on the other four hydrofacies; therefore the paleosols were modeled separately and later overprinted on the simulated geology of each sequence. Assembly of conditional simulation results for each sequence together with the paleosols resulted in a nonstationary, multiscale model of heterogeneity [*Weissmann and Fogg*, 1999].

[15] The three-dimensional cell size of 100 m, 200 m, and 0.5 m in the depositional strike, depositional dip, and vertical directions, respectively, was selected using two criteria: (1) these lengths are significantly less than the mean lengths of each facies [*Weissmann and Fogg*, 1999] and (2) sufficiently large to avoid computational limitations during groundwater flow simulation. The overall dimensions of the simulated region are 6300 × 15,000 × 100.5 m in the depositional strike, depositional dip, and vertical directions, respectively.

[16] A series of 10 equally probable realizations were developed for this study. Figure 3 shows one realization of the Kings River Alluvial Fan hydrostratigraphy used in the groundwater flow and random walk particle tracking simulations described in the following sections.

4.2. Groundwater Flow Simulation

[17] A three-dimensional steady state modeling approach was used to simulate groundwater flow conditions in the aquifer with the finite difference code MODFLOW-96 [*McDonald and Harbaugh*, 1988; *Harbaugh and McDonald*, 1996]. The study area was discretized into approximately 950,000 cells within a simulation grid of overall dimensions of 6300 × 15,000 × 100.5 m in the depositional strike, depositional dip, and vertical directions, respectively, from the geostatistical realizations. The constant cell dimensions from the geostatistical realizations (100 × 200 × 0.5 m) were used for the numerical flow simulation grid.

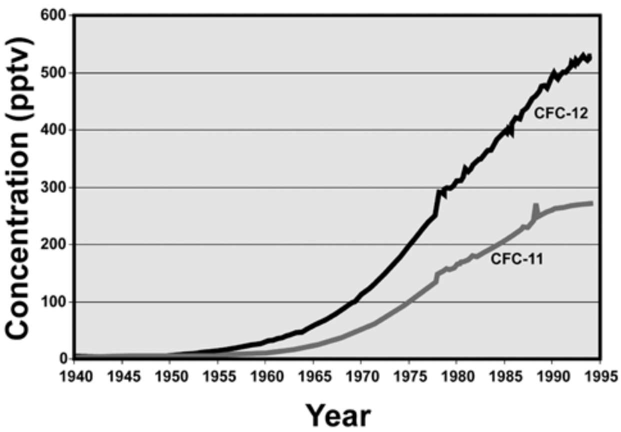

Figure 2. Atmospheric concentrations of CFC-11 and CFC-12 since 1940. Concentration values between 1940 and 1977 are from *Busenberg et al.* [1993]. Concentration values between 1977 and 1994 were taken from the NOAA/CMDL Niwot Ridge Station, Colorado, as reported by *Elkins et al.* [1999].

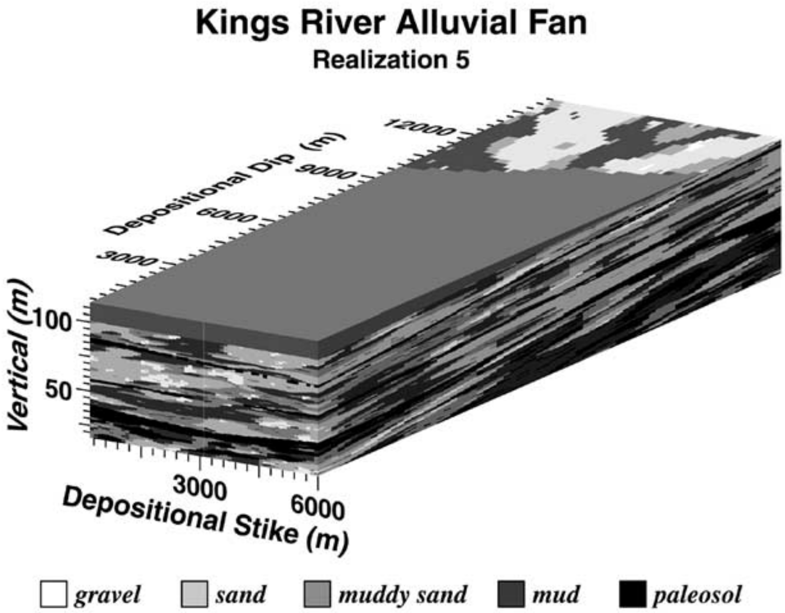

Figure 3. Geostatistical realization 5 from the Kings River Alluvial Fan system.

[18] Hydraulic conductivity (K) was assigned to each cell based on the simulated hydrofacies determined by the geostatistical realization for each scenario. Values used for K of each hydrofacies (Table 2) were estimated from pumping test, slug test, laboratory core measurements, and literature estimates for similar lithologies [*Burow et al.*, 1999; *Weissmann*, 1999].

[19] General head boundary conditions [*McDonald and Harbaugh*, 1988] were used in the modeling to simulate inflow and outflow through the lateral and basal boundaries of the model. Hydraulic head values were defined for these boundaries using a general gradient of 0.002 parallel to the stratigraphic dip direction. Minor adjustments to the boundary head values were made along the stratigraphic dip boundaries to account for local gradient variability reported in water-elevation maps of the study area [*Fresno Irrigation District*, 1993]. No vertical gradient was assigned because measured vertical gradients in the area are small and variable in both upward and downward directions [*Burow et al.*, 1999]. The boundary condition for the top of the model represents constant recharge to the water table at a rate of 150 mm/year [*Burow et al.*, 1999].

[20] Groundwater pumpage was not included in the model for two reasons: (1) the previous characterization and modeling study of the same area [*Burow et al.*, 1999] showed no perturbations in hydraulic head suggestive of significant pumping influence on horizontal or vertical gradients and (2) wells in the area are not metered. The main effect of pumping would be to increase downward groundwater velocities, yet measured heads in the USGS well nests show no pervasive, downward gradients. Leakage out of the lower boundary of the model into deeper, finer-grained alluvial materials may, in fact, be compensating for lack of specified pumpage in the model. Effects of pumpage on general characteristics of the local groundwater age

Table 2. Hydraulic Conductivity Values Assigned to the Five Hydrofacies

Hydrofacies	K, m/s
Gravel	1×10^{-2}
Sand	1×10^{-3}
Muddy sand	1×10^{-5}
Mud	1×10^{-6}
Paleosol	1.3×10^{-7}

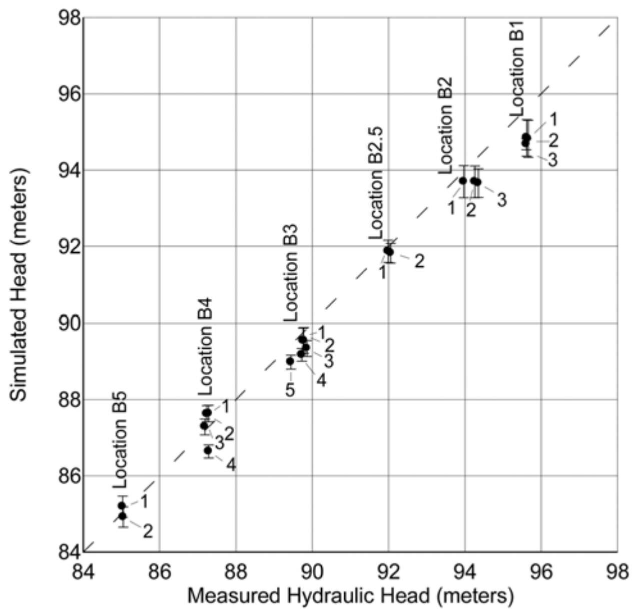

Figure 4. Measured hydraulic head [from *Burow et al.*, 1999] plotted against simulated mean hydraulic head at 20 monitoring well locations for the 10 realizations. Data points show mean results and error bars show the range of values from the 10 realizations.

distributions is expected to be minimal, but should be investigated in future studies.

[21] Figure 4 shows the simulated steady state hydraulic head at the midpoint of the 20 well screens plotted against measured head for these locations from *Burow et al.* [1999]. The average root-mean square error in head at the 20 measuring points for the 10 realizations is 2.41 meters, with r^2 value of near 1. Although modeled head values do not exactly match the measured values, simulated head values are close and the flow simulations preserved both the variability in vertical gradients and the overall observed horizontal gradients within the study area.

[22] The measured heads indicate no pervasive trend in vertical hydraulic gradients [*Burow et al.*, 1999], but rather shifting gradients from upward to downward, apparently due to flow around lenticular heterogeneities in three-dimensions. The simulated heads exhibit similar behavior, except at location B4, where the model tends to have downward gradients. In addition to flow simulations through the heterogeneous realizations, we simulated flow through a homogeneous K field for comparison purposes. Anisotropic values for K of 3.6×10^{-4} m/s, 4.3×10^{-4} m/s, and 1.7×10^{-5} m/s for the depositional strike, depositional dip, and vertical directions, respectively, were estimated from effective K properties of the geostatistical realizations. Effective K values in all three directions were determined through simulation of flow in the strike, dip, and vertical directions, as described by *Weissmann* [1999]. Similar boundary conditions to those previously described were used for these ‘‘homogeneous’’ simulations.

4.3. Particle Tracking Simulation Approach

[23] Transport simulations employed a random-walk particle tracking method (RWPM), described by *LaBolle et al.* [1998, 2000]. Application of the RWPM to simulate the forward-time advection-dispersion equation (ADE) models future particle distributions, with density representing solute concentration given knowledge of past distributions. When applied to simulate the backward-time ADE, the RWPM equation models past particle locations, with density representing probability given that future positions are known [*Uffink*, 1989; *Fogg et al.*, 1999; *Neupauer and Wilson*, 1999]. This procedure accurately models transport in a heterogeneous porous media, where sharp contrasts exist between lithologies. Details of the RWPM are described by *Uffink* [1989], *LaBolle et al.* [1996, 1998, 2000], and *LaBolle and Fogg* [2001], and are summarized in this section.

[24] In this approach, we assume that transport is described by an ADE of the form:

$$\frac{\partial nc}{\partial t} = -\frac{\partial v_i c}{\partial x_i} + \frac{\partial}{\partial x_i}\left(nD_{ij}\frac{\partial c}{\partial x_j}\right) \tag{2a}$$

$$D_{ij} = \left(\alpha_T|\mathrm{v}| + D'_d\right)\delta_{ij} + (\alpha_L - \alpha_T)v_i v_j/|\mathrm{v}| \tag{2b}$$

where c is the concentration, $\mathbf{v}$ is pore water velocity, D_{ij} is a dispersion tensor, n is effective porosity, D'_d is the molecular diffusivity in a porous media, δ_{ij} is the Dirac delta function, and α_L and α_T are longitudinal and transverse dispersivities, respectively. The dependent variable of (2a) can be replaced with the conditional probability $p(x,t|s,y)$, where $c(x,t) = p(\mathrm{x,t}) = p(x,t|s,y)p(s,y)$ with initial condition [*Arnold*, 1974]

$$\lim_{t\to s} p(x,t|y,s) = \delta(x-y) \tag{3}$$

where x is the final location of the particle at time t and y is the initial location of the particle at time s. The form of the backward-time ADE for constant n is given by the adjoint of (2a) [*Arnold*, 1974]

$$\frac{\partial p}{\partial s} = v_i\frac{\partial p}{\partial y_i} - \frac{\partial}{\partial y_i}\left(D_{ij}\frac{\partial p}{\partial y_j}\right) \tag{4a}$$

$$\lim_{s\to t} p(x,t|y,s) = \delta(x-y) \tag{4b}$$

The diffusion operator is said to be self-adjoint because it takes the same form in both the forward and backward processes. The advection term is not self-adjoint unless the velocity is divergence free, in which case (4a) is equivalent to

$$\frac{\partial p}{\partial s} = -\frac{\partial v_i p}{\partial y_i} - \frac{\partial}{\partial y_i}\left(D_{ij}\frac{\partial p}{\partial y_j}\right) \tag{5}$$

In reference to the forward equation, p in the backward equation (5) is evolving in s from some final time to some initial time. If, for convenience, one reverses time in (5) such that positive values of $s - t$ represent evolution backward in time, the backward ADE (5) simply takes the form of the forward ADE (2a) with the velocity reversed. Both equations are based on conditional probabilities. The backward equation gives information on locations where contaminants may have originated in the past [*Uffink*, 1989; *Fogg et al.*, 1999]. This backward-time ADE was used to compute the ages of groundwater entering a monitoring well from travel time between the well screen and water table.

[25] Parameter values required for the RWPM include velocities calculated from the flow simulations, molecular diffusivities, effective porosity, and longitudinal and transverse dispersivities. The molecular diffusivity (D'_d) was estimated from

$$D'_d = \frac{D_d}{\tau^2} \tag{6}$$

where D_d is the diffusion coefficient in a liquid [*Grathwohl*, 1998] and τ is tortuosity defined as

$$\tau = \frac{L_e}{L} \tag{7}$$

where L_e is the flow path length and L is the straight-line distance [*Carman*, 1956; *Grathwohl*, 1998; *Fetter*, 1999]. Using $D_d = 2 \times 10^{-9}$ m^2/s, an average of diffusion coefficients for common ions cited by *Domenico and Schwartz* [1998], and a tortuosity of 1.7 [*Carman*, 1956], D'_d was estimated to be approximately 6.9×10^{-10} m^2/s. An effective porosity value of 0.33 was applied for these simulations.

[26] *LaBolle* [1999] and *LaBolle and Fogg* [2001] showed that simulation results are insensitive to the value

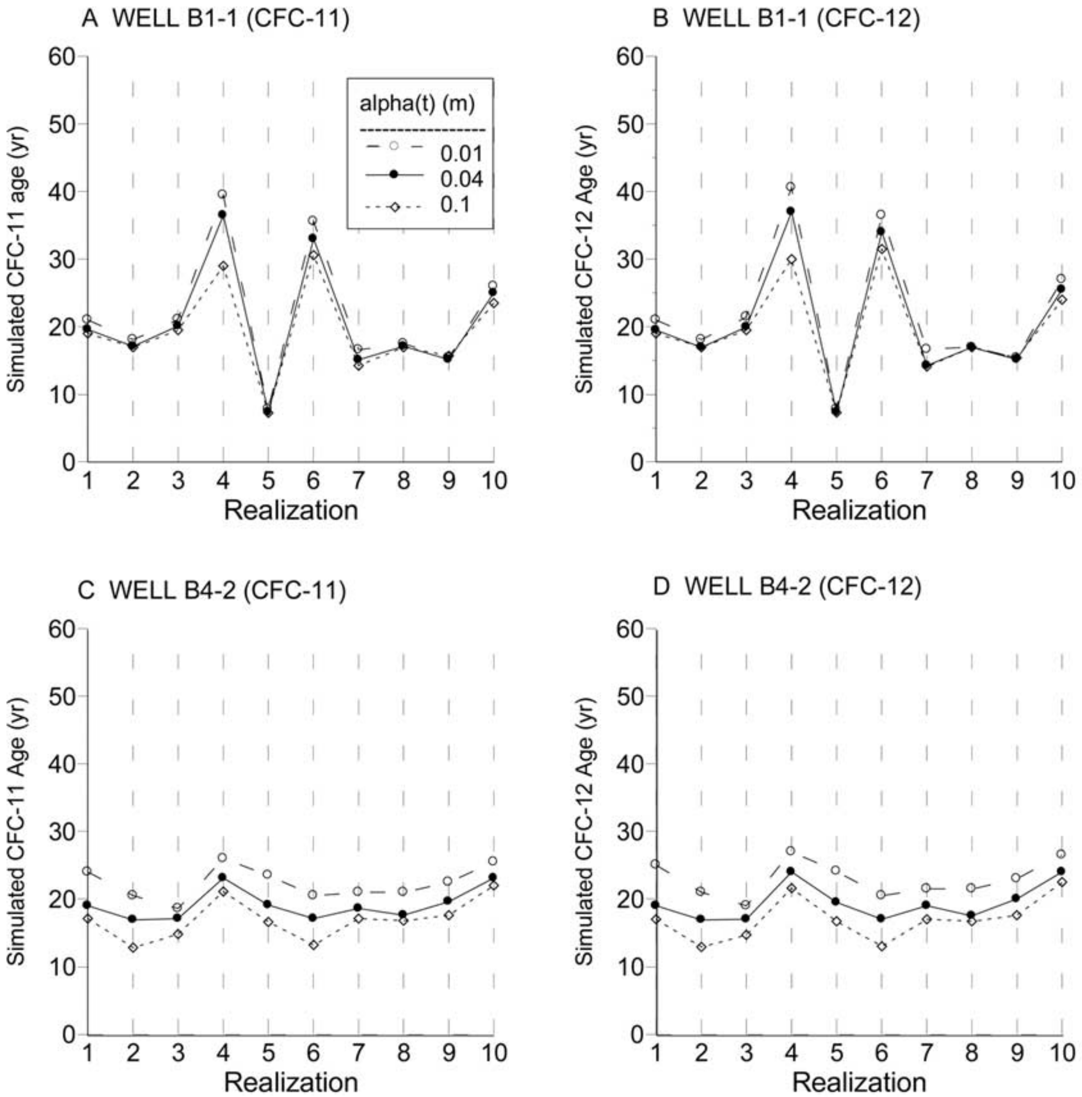

Figure 5. Sensitivity results of simulated CFC-based age estimates for 10 realizations using α_T = 0.01, 0.04, and 0.1 m for (a) CFC-11 ages at well B1-1, (b) CFC-12 ages at well B1-1, (c) CFC-11 ages at well B4-2, and (d) CFC-12 ages at well B4-2.

of local-scale α_L. This is true because the spreading due to longitudinal dispersion is insignificant compared to that caused by the hydrofacies-scale heterogeneity that is captured in the geostatistical simulations. Furthermore, the particular random walk particle algorithm that accurately balances mass across sharp interfaces runs faster when dispersivity is isotropic. Dispersivity was therefore treated as isotropic, with $\alpha_L = \alpha_T = 0.04$ m, a value similar in magnitude to reported transverse dispersivities at field sites similar to the scale of our cells [e.g., *Freyberg*, 1986; *Sudicky*, 1986; *Garabedian et al.*, 1991; *Farrell et al.*, 1994; *Mallants et al.*, 2000]. Because some uncertainty in this value exists, we conducted sensitivity runs with α set to 0.1 and 0.01 m. These simulations showed minimal effects on the results reported here (Figure 5).

4.4. Numerical Simulation of Groundwater Ages

[27] To assess the age distribution of water entering a well screen, we simulated backward-in-time release of 5000 particles from a discrete point located at the midpoint of 17 monitoring well screens at various depths in the study area (Table 1). Thus these simulations reflect groundwater age distributions at a discrete point in the aquifer. Particles in these simulations represent parcels of water of various ages. Using the steady state velocity fields along with the backward-time particle tracking approach previously described, the travel times for particles between these monitoring well screen midpoints and the water table were simulated for each of the 10 heterogeneous realizations and the homogeneous system. Because the backward-moving particles reach the water table at various times, the results of this procedure produces probability distribution of groundwater residence time at a well (Figure 6). Average residence time of groundwater reaching each well screen was calculated as the arithmetic mean of particle travel times (Table 1). Assuming that CFC atmospheric concentrations are the same as vadose zone air concentrations, and that water recharged at the water table is in equilibrium with this

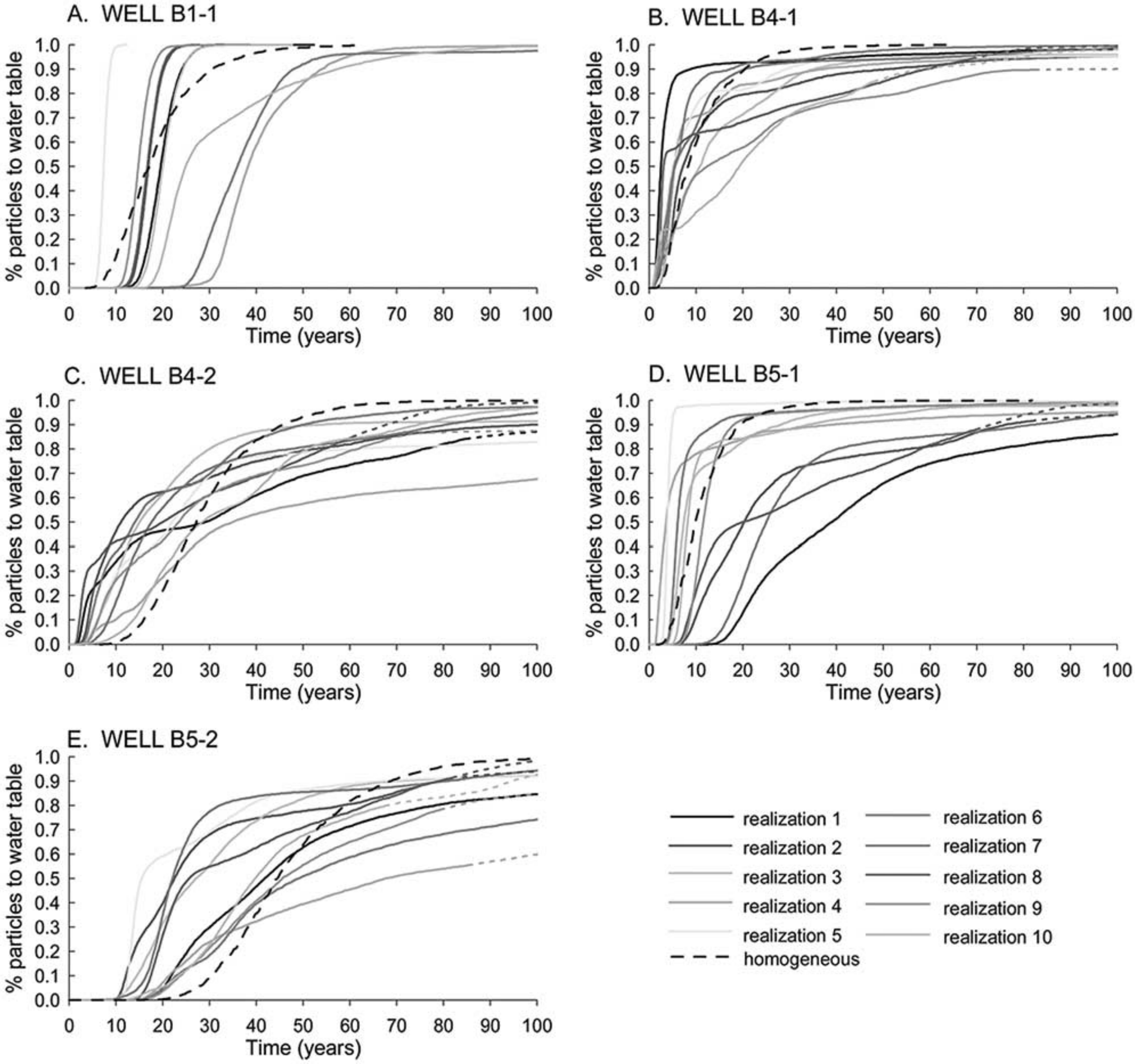

Figure 6. Simulated groundwater residence time distributions from the midpoint of selected well screens for each of the 10 realizations: (a) B1-1, (b) B4-1, (c) B4-2, (d) B5-1, and (e) B5-2. Dashed portions of the residence time distribution curves indicate that >10% of the particles required estimation of groundwater residence times using the depth versus residence time relationship of Figure 7. The thick dashed line shows simulated residence times assuming a homogeneous aquifer system. See color version of this figure at back of this issue.

concentration, this residence time should be equivalent to the CFC-based groundwater age.

[28] In several groundwater residence time simulations, a significant number of particles left the simulation through the upgradient boundary, never reaching the water table. Therefore we could not directly determine the residence time of those particles. Instead, we estimated their residence times by adding the estimated residence time based on a relationship between particle age and depth in the system. A depth versus residence time relationship was determined by averaging simulated groundwater residence time at several depths along the downgradient boundary (Figure 7). This depth versus residence time relationship carries much uncertainty for several reasons, including (1) upgradient geology may be significantly different since this will include the apex region of the alluvial fan and thus be dominated by a greater percentage of coarse-grained material; and (2) heterogeneity within a system, as shown by this research, causes significant variance around the depth versus residence time relationship at any depth. However, even with these potential sources of error for these particles, the overall influence of these particles is minimal for most wells and simulations (Figure 6) since these particles represent water with residence times >55 years and would thus contain no CFC. This assumption allows a means of estimating the full distribution of groundwater residence

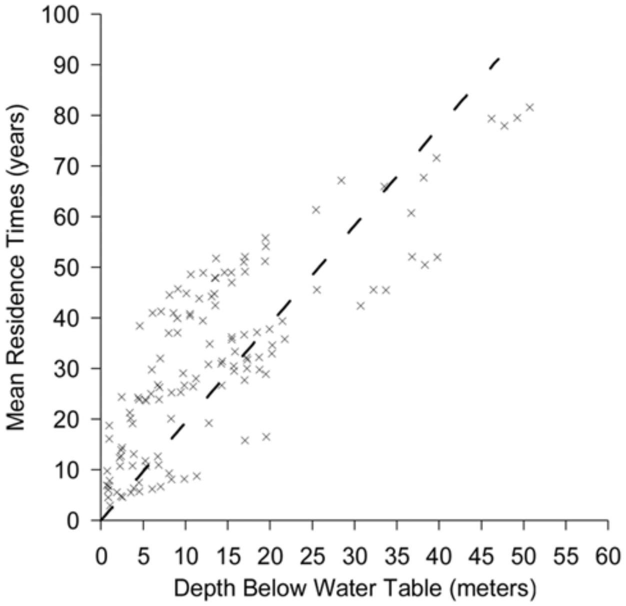

Figure 7. Estimated residence time versus depth relationship for the study area produced by estimating groundwater residence times for cells along the downgradient boundary of the study area.

time while maintaining a simulation space small enough to conduct the numerical experiments within a reasonable amount of time.

[29] Simulated CFC age dates were obtained by finding in Figure 2 the reported atmospheric CFC concentration values ($C_{CFC,p}$) corresponding to residence time of each particle. A total CFC concentration was estimated at each well using

$$C_{CFC} = \frac{\sum_{N_p} C_{CFC,p}}{N_p} \qquad (8)$$

where N_p is the number of particles. The resulting "simulated CFC-age" of groundwater was estimated by determining the year in Figure 2 that corresponds to this calculated CFC concentration at the well. In this application, we assume that the temperature at recharge depth remains constant over the length of time simulated in order to honor Henry's Law.

5. Results

5.1. General Observations

[30] Simulations of groundwater residence time distributions show that groundwater tapped by each well consists of a wide mix of water with various residence times, even though the particles were released from points in space rather than from the entire ~1.5-m-long well screens (Table 1 and Figure 6). A high degree of residence time distribution variability between the 10 different realizations was observed (Figure 6) and is also reflected in standard deviation of residence times computed for the various scenarios (Table 1). This variability reflects the uncertainty inherent to our conditional simulations of heterogeneity in the system. Even with this uncertainty, most curves associated with a well retain similar shape and breadth of age distribution.

[31] With the exception of Well B1-1 and Well B5-1, the broad age distributions and their asymmetry indicate that non-Fickian dispersion caused by facies-scale heterogeneity is significant. In the case of Wells B1-1, most simulated age results agree fairly closely with the normal age distribution produced under homogeneous conditions (Figure 6a). In the Well B1-1 simulations that display this normal distribution, the particles traveled primarily through one hydrofacies type with minor excursions into other hydrofacies. Simulations that produced a broader residence time distribution (e.g., realizations 4, 6, and 10) were the result of particles traveling through several hydrofacies between the well screen and water table. Similar responses were observed in simulations conducted from Well B5-1, where all but four of the simulations showed Fickian response related to transport through primarily one hydrofacies. Therefore the residence time distribution shape appears to be highly dependant on the degree of heterogeneity encountered by the tracer.

[32] Simulated CFC-based groundwater ages favorably match measured CFC-based groundwater ages reported by *Burow et al.* [1999] (Figure 8 and Table 1), with some anomalies that are explainable. Simulated CFC-based groundwater ages are generally slightly older than reported CFC ages, especially for ages from CFC-12. This may stem from the lack of regional pumping in our simulations that would potentially draw a greater proportion of younger water to the deeper screened intervals in the model.

5.2. Anomalies

[33] Only simulation results for wells B2-2, B2.5-2, B3-4, and B4-4 show noticeably greater CFC ages than those reported by *Burow et al.* [1999]. Sample measurement error, possibly caused by CFC contamination of the sample (thus producing an artificially young age result since concentrations would be increased), may explain some of these discrepancies. For example, the measured groundwater age from CFC-12 for Well B4-4 is 29.5 yrs, which is inconsistent with the lack of detectable tritium [*Burow et al.*, 1999] and the depth of this well. Furthermore, 1,2-Dibromo-3-Chloropropane (DBCP) and nitrate concentrations, contaminants determined to be associated with younger groundwater [*Burow et al.*, 1999], along with specific conductance in groundwater sampled from this well suggest that the water from this well is older than 29.5 yrs [*Burow et al.*, 1999]. Finally, each of the 10 simulated ages from the 10 geostatistical realizations for Well B4-4 is older than measured value (Table 1). The discrepancy between measured and modeled age may also be related to a problem with the geostatistical representation of heterogeneity at that location since simulated vertical gradients at location B4 were consistently downward in contrast to the measured gradients at that location. *Burow et al.* [1999], however, noted that some of the samples from the study site were probably contaminated with CFC-11 or CFC-12.

[34] Discrepancies between our simulated groundwater ages and observed CFC ages were also noted in the CFC-11 age dates for well B3-4. The simulated CFC-11 groundwater age of 38.1 years is greater than the observed 29 years for CFC-11 in this well. However, the simulated CFC-12

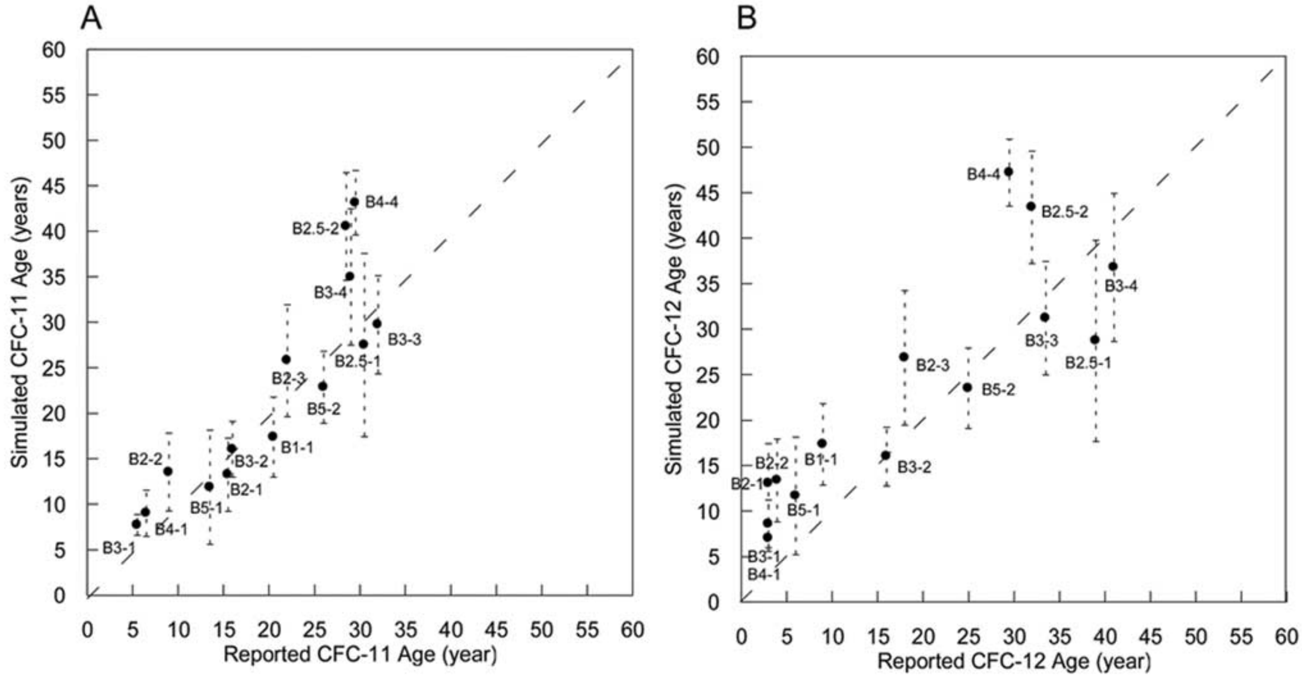

Figure 8. Mean simulated CFC-based groundwater ages plotted against measured CFC-based groundwater ages reported by *Burow et al.* [1999] for (a) CFC-11 and (b) CFC-12. Ranges around the mean show the range of simulated CFC-based ages within the 10 realizations.

age of 40.5 favorably matches the observed 41-year CFC-12 age date. Additionally, the age trend between Wells B3-1, B3-2, B3-3, B3-4, and B3-5 show the expected tendency of increasing age with depth at the B3 location, and the measured B3-4 age date appears to be too young to fit this trend. *Burow et al.* [1999] indicate that CFC-11 is more susceptible to microbial transformation, however this was not thought to be a dominant factor affecting CFC-11 concentrations due to low organic content and well-oxygenated groundwater. Additionally, CFC-12 is less likely than CFC-11 to be contaminated by sampling [*Bartolino*, 1997]. *Burow et al.* [1999] assigned a recharge date to this older value from CFC-12 data. Our results suggest this older age date is more appropriate.

[35] Finally, the measured CFC-11 age of water in Well B2-2 (8 years) is less than that observed in Well B2-1 (16 years), indicating that the deeper aquifer holds younger water at the B2 location. This same phenomenon was reported for measured ages at Well B2.5-1 and Well B2.5-2. None of the simulations within the 10 realizations reflected this reversed groundwater age, thus simulated CFC-11 and CFC-12 ages are greater than the measured values. These age discrepancies may be either due to contamination of the CFC samples, thus producing a younger age date, or presence of short-circuit flow paths close to wells B2 and B2.5 in the field but not in the model.

5.3. Simulated CFC Age and the Mean Age

[36] In order to assess whether CFC ages reflect the mean groundwater age of a sample in a heterogeneous aquifer, we compared the simulated CFC age results to the simulated overall mean groundwater residence time at a well (Figure 9). Simulated CFC-based age dates are consistently lower than the actual mean residence time of the simulated samples. This is because the CFC age dating cannot differentiate ages of water older than 55 years. Therefore it appears that CFC-based ages do not typically reflect the mean age of the groundwater in a typically heterogeneous alluvial system. This result supports similar observations by *Varni and Carrera* [1998], *Tompson et al.* [1999], and *Bethke and Johnson* [2002].

6. Discussion and Conclusions

6.1. Residence Time Dispersion and Mean Age

[37] The simulated residence time distributions (Figure 6 and Table 1) exhibit substantial variance and positive skewness (tailing), with some water "samples" containing water ages ranging from 5–10 yrs to >100 yrs. The resulting CFC age dates represent biased sampling of these distributions, thereby underestimating actual mean age. The bias increases with mean age (Figure 9), owing to the growth in residence time variance with mean age.

[38] The substantial variance of residence times is caused primarily by spatially variable groundwater velocities that are inherent to the three-dimensional, heterogeneous alluvial system (Figure 3). Mixing of residence times along the sampling interval is not a significant factor here because the well screens are short (~1.5 m). The tailing behavior is caused by presence of very low-K units (paleosols and muds) in which relatively slow advection and diffusion dominate transport [*LaBolle and Fogg*, 2001].

[39] The results clearly illustrate how representation of groundwater age by a single date can be misleading. Only when the flow paths are relatively short, as in the case of wells B1-1 and B5-1, does the groundwater residence time distribution approximate a normal distribution with small variance for some realizations of the heterogeneity

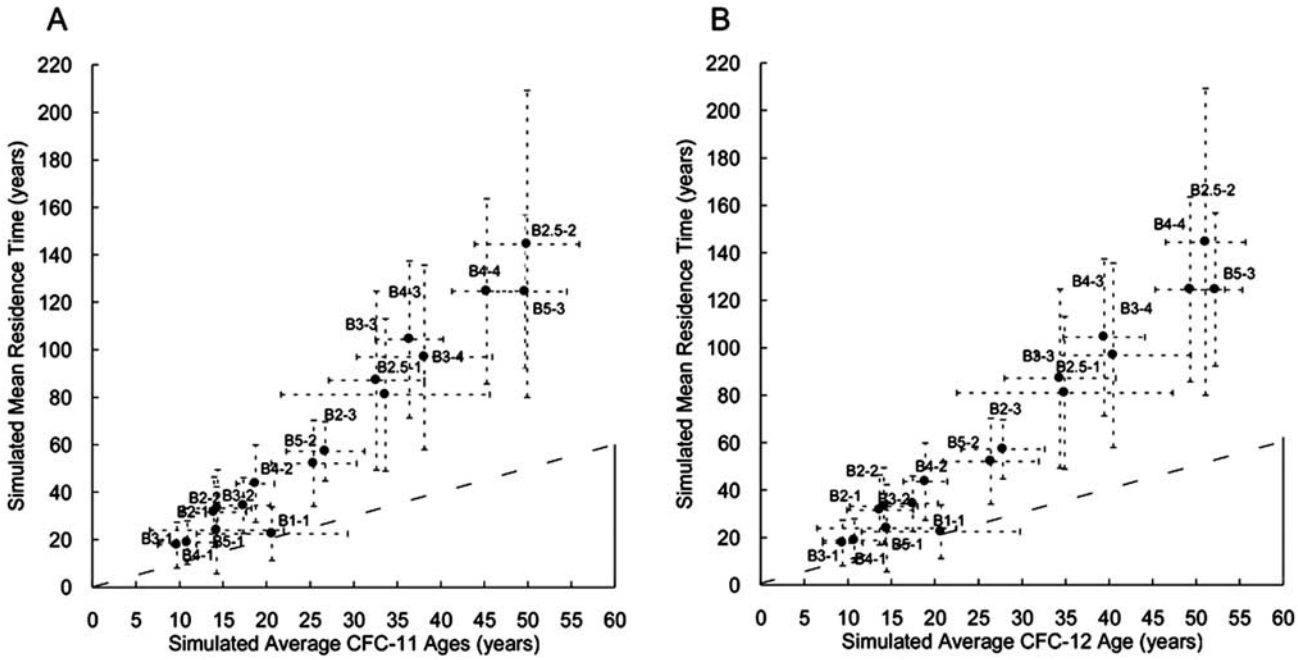

Figure 9. Simulated mean groundwater residence times plotted against mean simulated CFC-based groundwater ages from the 10 realizations for (a) CFC-11 and (b) CFC-12. The error bars show the ranges of simulated groundwater residence times and CFC ages for each of the 10 realizations.

(Figure 6), and, thus only in these cases will simulated CFC ages reasonably correspond to the mean groundwater ages.

[40] Even if the age date technique is capable of unbiased sampling, interpretation of the field measured ages can be confounded by the broad range of ages within the sample. For example, the notion that an age date of, say, 40 years is always indicative of less vulnerability to recent, anthropogenic contamination than an age of 20 years is flawed. If the source of contamination began 30 years ago, depending on the groundwater residence time distributions, the 40-year water could contain as much or more water that is younger than 30 years as the 20-year water. Furthermore, if the age of a water is estimated to be very old (e.g., thousands of years), the tailing phenomenon could have the effect of including very young water in the same sample. Such extreme tailing of residence times is consistent with *Bethke and Johnson*'s [2002] analysis, which indicates that diffusion induced tailing can cause mixing of young and very old waters locally within an aquifer. Even without diffusion-induced tailing, it is expected that regional groundwater flow systems would commonly include both old water that has traveled great distances and younger water that enters the regional aquifer system via leakage or preferential, vertical flow through discontinuities in confining beds. Groundwater pumpage would accelerate the downward flow of relatively young water and enhance the mixing of disparate groundwater ages. Future research should be directed at assessing potential age dispersion and age sampling bias for a variety of environmental tracers and aquifer systems.

[41] The hypothesis that CFC-based age dates represent an average age appears to be with little merit for all but the most simple flow systems involving relatively young (<55 years) groundwater. Several writers have indicated that this may be the case [e.g., *Walker and Cook*, 1991; *Mazor and Nativ*, 1992; *Goode*, 1996; *Varni and Carrera*, 1998; *Fogg et al.*, 1999; *Tompson et al.*, 1999; *Bethke and Johnson*, 2002]. The present study models the full residence time distributions, thereby quantifying and elucidating the potential errors and ambiguities for a highly characterized, heterogeneous system.

[42] The bias problem in age dating techniques that preferentially detect the relatively young waters (e.g., CFC and tritium-based methods) could be evaluated more directly with an age dating technique that works well for groundwater ages in the intermediate range (e.g.,<1000 yr). While techniques exist for dating much older waters (e.g., ^{14}C, ^{36}Cl), no techniques are available for dating the intermediate range. Clearly, intermediate-age dating techniques are needed.

6.2. Implications for Groundwater Quality Sustainability

[43] *Fogg et al.* [1999] simulated dispersion of groundwater residence times in a regional-scale (~45 km) analysis of a portion of the Salinas Valley. Similarly, their results indicated significant dispersion of groundwater residence time within simulated water "samples," with residence times in individual "samples" often ranging from 10 to >500 yrs. Their model indicated such mixing over most of the Salinas Valley study area, which receives nonpoint source contaminant (nitrate) loading primarily from fertilizer sources. *Fogg et al.* [1999] pointed out a weighty implication of such extreme mixing - that wells exhibiting contamination today in alluvial aquifers similar to the Salinas Valley (e.g., San Joaquin Valley, basin and range aquifers, Gulf Coast aquifers, etc.) commonly derive only a fraction of their production from the waters that are young

enough to be contaminated. Thus, if the sources of recalcitrant contaminants (nitrates and salinity in the Salinas Valley case) have not diminished appreciably since their introduction starting in the 1940s and 1950s, one can expect steadily declining water quality many decades into the future, even if the sources are reduced or eliminated today. In other words, if the groundwater contamination that we see today in the Salinas Valley is due only to contamination of that fraction of the water pumped by wells that has been exposed to anthropogenic contaminants, this fraction will increase for many decades or centuries into the future, leading to gradually rising concentrations at the wellhead. Because of the slow response time of basin-scale groundwater quality in deep, alluvial systems like the Salinas and San Joaquin Valleys, this degradation would be virtually irreversible on the scale of decades to centuries.

[44] Such a scenario has played out with respect to pesticide contamination in the Kings River fan aquifer [*Burow et al.*, 1999], where use of the chemical DBCP circa 1950–1972 has left a legacy of contamination in wells up through the present. DBCP concentrations in this system appear to have been declining in recent years owing to chemical transformation and/or recycling of groundwater through pumpage and reapplication for irrigation. The average groundwater residence times in this comparatively thin, coarse-grained system are young relative to many other portions of the San Joaquin Valley and many other alluvial basins. Thus the contamination and subsequent recovery of groundwater quality in the Kings River fan aquifer system within 30 to >50 yrs can be considered relatively fast. It is likely that sources of contamination in finer-grained aquifer systems (e.g., west side of the San Joaquin Valley) where the time constants for transport are much greater have not yet had full impact on deeper aquifers.

[45] The present study was conducted in part to investigate the age dispersion processes in greater detail, with much more detailed hydrostratigraphic characterization and a more carefully calibrated model, including some validation with measured CFC-based ages. Much like the results of *Fogg et al.* [1999], we find significant dispersion of ages within individual water "samples," albeit for less regional length and timescales. Thus the work of this study reinforces the conclusions of *Fogg et al.* [1999] regarding groundwater sustainability in the presence of nonpoint source, recalcitrant contaminants.

[46] Such trends in groundwater quality can only be detected through very long-term monitoring, which is typically rare. Although groundwater hydrologists and managers intuitively know that groundwater quality changes can be very slow, there is a strong tendency to stop monitoring water quality when certain chemical constituents show little variation for several months or years. Although data on long-term water quality trends are rare, some of the available data show an alarming consistency with the above hypothesis regarding long-term degradation of groundwater quality. For example, *Dubrovsky et al.* [1998, p. 17] show average groundwater nitrate concentrations rising steadily from the 1950s through the 1980s and 1990s in the eastern San Joaquin Valley. Such data together with our improved understanding of groundwater residence time dispersion suggest that in many basins we may presently be viewing only the "early breakthrough" of this centuries long "tracer test." Continued long-term water quality monitoring and process investigation are therefore paramount.

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References

Arnold, L., *Stochastic Differential Equations: Theory and Applications*, 228 pp., John Wiley, New York, 1974.

Bartolino, J. R., Chlorofluorocarbon and tritium age determination of ground-water recharge in the Ryan Flat subbasin, Trans-Pecos, Texas, *U.S. Geol. Surv. Water Res. Invest.*, *96-4245*, 29 pp., 1997.

Bethke, C. M., and T. M. Johnson, Paradox of groundwater age, *Geology*, *30*(2), 107–110, 2002.

Burow, K. R., G. S. Weissmann, R. D. Miller, and G. Placzek, Hydrogeologic facies characterization of an alluvial fan near Fresno, California, using geophysical techniques, *U.S. Geol. Surv. Open File Rep.*, *97–46*, 15 pp., 1997.

Burow, K. R., S. Y. Panshin, N. M. Dubrovsky, D. VanBrocklin, and G. E. Fogg, Evaluation of processes affecting 1,2-dibromo-3-chloropropane (DBCP) concentrations in ground water in the eastern San Joaquin Valley, California: Analysis of chemical data and ground-water flow and transport simulations, *U.S. Geol. Surv. Water Resour. Invest.*, *99–4059*, 1999.

Busenberg, E., and L. N. Plummer, Use of chlorofluorocarbons (CCl_3F and CCl_2F_2) as hydrologic tracers and age-dating tools: The alluvium and terrace system of central Oklahoma, *Water Resour. Res.*, *28*, 2257–2283, 1992.

Busenberg, E., E. P. Weeks, L. N. Plummer, and R. C. Bartholomay, Age dating ground water by use of chlorofluorocarbons (CCl_3F and CCl_2F_2), and distribution of chlorofluorocarbons in the unsaturated zone, Snake River Plain Aquifer, Idaho National Engineering Laboratory, Idaho, *U.S. Geol. Surv. Water Resour. Invest. Rep.*, *93–4054*, 47 pp., 1993.

Carle, S. F., and G. E. Fogg, Transition probability-based indicator geostatistics, *Math. Geol.*, *28*, 453–476, 1996.

Carle, S. F., E. M. LaBolle, G. S. Weissmann, D. Van Brocklin, and G. E. Fogg, Conditional simulation of hydrofacies architecture: A transition probability/Markov approach, in *Hydrogeologic Models of Sedimentary Aquifers, Concepts in Hydrogeol. and Environ. Geol.*, vol. 1, edited by G. S. Fraser and J. M. Davis, *Spec. Publ. SEPM Soc. Sediment. Geol.*, 147–170, 1998.

Carman, P. C., *Flow of Gases Through Porous Media*, 182 pp., Academic, San Diego, Calif., 1956.

Cook, P. G., D. K. Solomon, L. N. Plummer, E. Busenberg, and S. L. Schiff, Chlorofluorocarbons as tracers of groundwater transport processes in a shallow, silty sand aquifer, *Water Resour. Res.*, *31*, 425–434, 1995.

Domenico, P. A., and F. W. Schwartz, *Physical and Chemical Hydrogeology*, 2nd ed., John Wiley, New York, 1998.

Dubrovsky, N. M., C. R. Kratzer, L. R. Brown, J. M. Gronberg, and K. R. Burow, Water quality in the San Joaquin-Tulare Basins, California, 1992–95, *U.S. Geol. Surv. Circ.*, *1159*, 38 pp., 1998.

Dunkle, S. A., L. N. Plummer, E. Busenberg, P. J. Phillips, J. M. Denver, P. A. Hamilton, R. L. Michel, and T. B. Coplen, Chlorofluorocarbons (CCl_3F and CCl_2F_2) as dating tools and hydrologic tracers in shallow groundwater of the Delmarva Peninsula, Atlantic Coastal Plain, United States, *Water Resour. Res.*, *29*, 3837–3860, 1993.

Elkins, J. W., T. M. Thompson, T. H. Swanson, J. H. Butler, B. D. Hall, S. O. Cummings, D. A. Fisher, and A. G. Raffo, Decrease in the growth rates of atmospheric chlorofluorocarbons 11 and 12, *Nature*, *364*, 780–783, 1993.

Elkins, J. W., T. M. Thompson, J. H. Butler, and R. C. Myers, Atmospheric Measurements of CFC-11 and CFC-12 from the NOAA/CMDL Flask Program, Natl. Oceanic and Atmos. Admin, Silver Spring, Md., 1999. (Available at ftp://ftp.cmdl.noaa.gov/noah/cfcs/)

Farrell, D. A., A. D. Woodbury, and E. A. Sudicky, The 1978 Borden tracer experiment: Analysis of the spatial moments, *Water Resour. Res.*, *30*, 3213–3223, 1994.

Fetter, C. W., *Applied Hydrogeology*, 4th ed., 598 pp., Macmillan, New York, 1999.

Fogg, G. E., E. M. LaBolle, and G. S. Weissmann, Groundwater vulnerability assessment: hydrogeologic perspective and example from Salinas Valley, California, in *Assessment of Non-Point Source Pollution in the Vadose Zone*, *Geophys. Monogr. Ser.*, vol. 108, edited by D. L. Corwin, K. Loague, and T. R. Ellsworth, pp. 45–61, AGU, Washington, D. C., 1999.

Fresno Irrigation District, Ground-water Elevation Map, January, 1993, scale 1 inch = 2.5 miles, Fresno, Calif., 1993.

Freyburg, D. L., A natural gradient experiment on solute transport in a sand aquifer, 2, Spatial moments and the advection and dispersion of nonreactive tracers, *Water Resour. Res.*, *22*(13), 2031–2046, 1986.

Garabedian, S. P., D. R. LeBlanc, L. W. Gelhar, and M. A. Celia, Large-scale natural gradient tracer test in sand and gravel. Cape Cod, Massachusetts, 2, Analysis of spatial moments for a nonreactive tracer, *Water Resour. Res.*, *27*, 911–924, 1991.

Goode, D. J., Direct simulation of groundwater age, *Water Resour. Res.*, *32*, 289–296, 1996.

Grathwohl, P., *Diffusion in Natural Porous Media: Contaminant Transport, Sorption/Desorption and Dissolution Kinetics*, 207 pp., Kluwer Acad., Norwell, Mass., 1998.

Harbaugh, A. W., and M. G. McDonald, User's documentation for MODFLOW-96, an update to the U.S. Geological Survey Modular Finite_Difference Ground-Water Flow Model, *U.S. Geol. Surv. Open File Rep.*, *96–485*, 1996.

Hackley, K. C., C. L. Liu, and D. D. Coleman, C-14 dating of groundwater containing microbial CH_4, *Radiocarbon*, *34*(3), 686–695, 1992.

Hinkle, S. R., and D. T. Snyder, Comparison of Chlorofluorocarbon age dating with particle tracking results of a regional ground-water flow model of the Portland Basin, Oregon and Washington, *U.S. Geol. Surv. Water Supply Pap.*, *2483*, 1997.

Johnston, C. T., P. G. Cook, S. K. Frape, L. N. Plummer, E. Busenberg, and R. J. Blackport, Ground water age and nitrate distribution within a glacial aquifer beneath a thick unsaturated zone, *Ground Water*, *36*, 171–180, 1998.

LaBolle, E. M., Theory and simulation of diffusion processes in porous media, Ph.D. dissertation, 202 pp., Univ. of Calif., Davis, 1999.

LaBolle, E. M., and G. E. Fogg, Role of molecular diffusion in contaminant migration and recovery in an alluvial aquifer system, *Transp. Porous Media*, *42*, 155–179, 2001.

LaBolle, E. M., G. E. Fogg, and A. F. B. Tompson, Random-walk simulation of transport in heterogeneous porous media: Local mass-conservation problem and implementation methods, *Water Resour. Res.*, *32*, 583–593, 1996.

LaBolle, E. M., J. Quastel, and G. E. Fogg, Diffusion theory for transport in porous media: Transition probability densities of diffusion processes corresponding to advection-dispersion equations, *Water Resour. Res.*, *34*, 1685–1693, 1998.

LaBolle, E. M., J. Quastel, G. E. Fogg, and J. Gravner, Diffusion processes in composite porous media and their numerical integration by random walks: Generalized stochastic differential equations with discontinuous coefficients, *Water Reour. Res.*, *36*(3), 651–662, 2000.

Mallants, E., A. Espino, M. Van Hoorick, J. Feyen, N. Vandenberghe, and W. Loy, Dispersivity estimates from a tracer experiment in a sandy aquifer, *Ground Water*, *38*(2), 304–310, 2000.

Mazor, E., and R. Nativ, Hydraulic calculation of groundwater flow velocity and age examination of the basic premises, *J. Hydrol.*, *138*, 211–222, 1992.

McDonald, M. G., and A. W. Harbaugh, A modular three-dimensional finite difference ground-water flow model, *U.S. Geol. Surv. Tech. Water Resour. Invest.*, *book 6*(chap. A1), 1988.

Neupauer, R. M., and J. L. Wilson, Adjoint method for obtaining backward-in-time location and travel time probabilities of a conservative groundwater contaminant, *Water Resour. Res.*, *35*(11), 3389–3398, 1999.

Plummer, L. N., and C. L. Sprinkle, Radiocarbon dating of dissolved inorganic carbon in groundwater from confined parts of the upper Floridan aquifer, Florida, USA, *Hydrogeol. J.*, *9*(2), 127–150, 2001.

Plummer, L. N., R. L. Michel, E. M. Thurman, and P. D. Glynn, Environmental tracers for age dating young ground water, in *Regional Ground-Water Quality*, edited by W. M. Alley, pp. 255–294, Van Nostrand Reinhold, New York, 1993.

Reilly, T. E., L. N. Plummer, P. J. Phillips, and E. Busenberg, The use of simulation and multiple environmental tracers to quantify groundwater flow in a shallow aquifer, *Water Resour. Res.*, *30*, 421–433, 1994.

Solomon, D. K., and E. A. Sudicky, Tritium and helium-3 isotope ratios for direct estimation of spatial variations in groundwater recharge, *Water Resour. Res.*, *27*, 2309–2319, 1991.

Solomon, D. K., R. J. Poreda, S. L. Schiff, and J. A. Cherry, Tritium and helium-3 as groundwater age tracers in the Borden Aquifer, *Water Resour. Res.*, *28*, 741–755, 1992.

Sudicky, E. A., A natural gradient experiment on solute transport in a sand aquifer: Spatial variability of hydraulic conductivity and its role in the dispersion process, *Water Resour. Res.*, *22*(13), 2069–2082, 1986.

Tompson, A. F. B., S. F. Carle, N. D. Rosenberg, and R. M. Maxwell, Analysis of groundwater migration from artificial recharge in a large urban aquifer: A simulation perspective, *Water Resour. Res.*, *35*(10), 2981–2998, 1999.

Uffink, G. J. M., Application of Kolmogorov's backward equation in random walk simulations of groundwater contaminant transport, in *Contaminant Transport in Groundwater: Proceedings of the International Symposium on Contaminant Transport in Groundwater*, edited by H. E. Kobus and W. Kinzelbach, pp. 283–298, A. A. Balkema, Brookfield, Vt., 1989.

Varni, M., and J. Carrera, Simulation of groundwater age distributions, *Water Resour. Res.*, *34*, 3271–3281, 1998.

Walker, G. R., and P. G. Cook, The importance of considering diffusion when using carbon-14 to estimate groundwater recharge to an unconfined aquifer, *J. Hydrol.*, *128*, 41–48, 1991.

Warner, M. J., and R. F. Weiss, Solubilities of chlorofluorocarbons 11 and 12 in water and seawater, *Deep Sea Res.*, *32*, 1485–1497, 1985.

Weissmann, G. S., Toward new models of subsurface heterogeneity: An alluvial fan sequence stratigraphic framework with transition probability geostatistics, Ph.D. dissertation, Univ. of Calif., Davis, 1999.

Weissmann, G. S., and G. E. Fogg, Multi-scale alluvial fan heterogeneity modeled with transition probability geostatistics in a sequence stratigraphic framework, *J. Hydrol.*, *226*, 48–65, 1999.

Weissmann, G. S., S. F. Carle, and G. E. Fogg, Three-dimensional hydrofacies modeling based on soil surveys and transition probability geostatistics, *Water Resour. Res.*, *35*, 1761–1770, 1999.

Weissmann, G. S., J. F. Mount, and G. E. Fogg, Glacially-driven cycles in accumulation and sequence stratigraphy of a stream-dominated alluvial fan, San Joaquin Valley, California, *J. Sediment. Res.*, *72*(2), 240–251, 2002.

G. Fogg, E. M. LaBolle, and Y. Zhang, Hydrologic Sciences, University of California, One Shields Avenue, Davis, CA 95616, USA.

G. Weissmann, Department of Geological Sciences, Michigan State University, 206 Natural Science Building, East Lansing, MI 48824–1115, USA. (weissman@msu.edu)

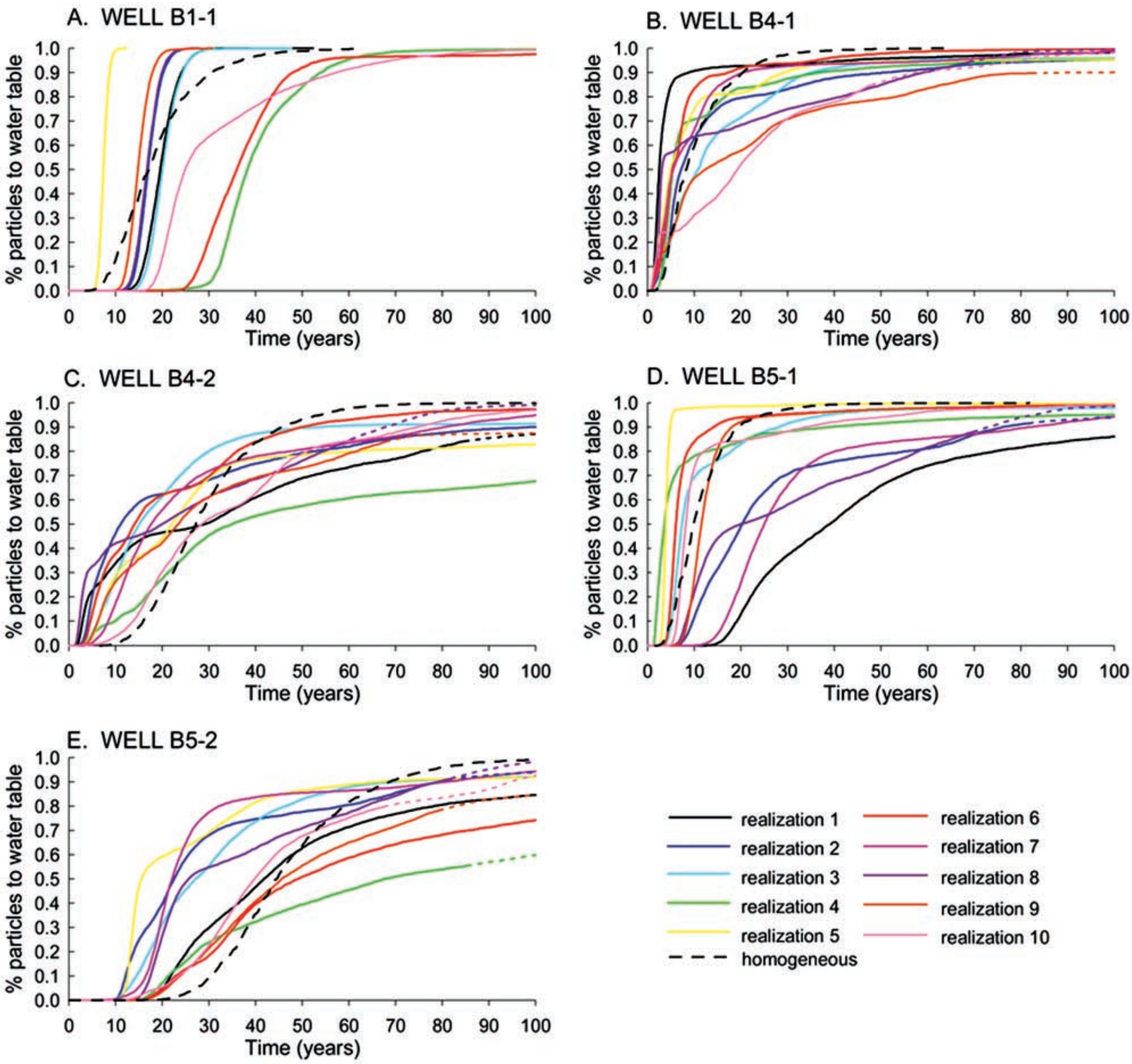

Figure 6. Simulated groundwater residence time distributions from the midpoint of selected well screens for each of the 10 realizations: (a) B1-1, (b) B4-1, (c) B4-2, (d) B5-1, and (e) B5-2. Dashed portions of the residence time distribution curves indicate that >10% of the particles required estimation of groundwater residence times using the depth versus residence time relationship of Figure 7. The thick dashed line shows simulated residence times assuming a homogeneous aquifer system.