

Parametric Study on Tracer Tests with Kinetic Exchange Processes for Georeservoir Characterization

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ABSTRACT

Artificial-tracer tests can provide a useful tool for evaluating kinetic exchange processes in dual-porosity media, if certain requirements are met. Simulated tracer signals ('breakthrough curves', BTC) for five typical kinetic-exchange process scenarios (roughly covering the broad range of solute partitioning processes that may become relevant for deep-georeservoir characterization) are seen to respond to variations of kinetic exchange parameter values in a monotonous, and fairly sensitive manner. In 'flow-storage repartition' terms (a tool proposed by Shook 2003 for characterizing what Shook deems as 'reservoir geometry'), apparent FSR shapes derived from simulated tracer BTCs are seen, as well, to largely follow the variations in kinetic-exchange parameters. Some of these FSR simulation findings seem somewhat surprising at first sight, but become understandable by comparing the degree of imbalance between fixation and release rates to the degree of deviation from the diagonal shape corresponding to the uniform plug-flow system. BTC simulation findings include: peak height is roughly determined by the fixation rate α , and it decreases with increasing α ; the tailing decrease rate is roughly determined by the release rate β ; peak arrival times and peak interval durations (before the onset of late tailings) depend on both α and β ; the tailing decrease rate gets accelerated with increasing β ; the higher the value of β , the lower the late tailings, and the later the onset of late tailings for a given value of α . For the particular case of matrix diffusion processes in their first-order approximation, the higher the fluid-rock interface area density, the later and lower the peak value, the higher and faster the mid-term tailings, but the lower and longer-lasting the late tailing levels. Apparent FSR findings include: for a given α value, the flow capacity to any given storage capacity decreases with increasing β (which seems surprising at first sight); in other words, with decreasing fixation-vs-release imbalance (with β slower than α), the apparent FSR shape approaches the diagonal shape of a uniform plug-flow system; for a given β value, increasing the value of α will reduce the flow capacity at high storage values, while raising the flow capacity at low storage values. For the particular case of matrix diffusion processes in their first-order approximation, the higher the fluid-rock interface area density, the lower the flow capacity for any given storage capacity, and the closer the apparent FSR shape will approach the diagonal. The latter looks surprising at first sight, but can be understood from the fact that, when α approximately equals β , both much faster than $1/MRT$, the matrix diffusion process approaches an equilibrium-retardation process (towards which FSR shapes are insensitive), while on the other hand the first-order approximation suggested by various authors becomes inadequate for describing matrix diffusion processes.

1. INTRODUCTION

Georeservoirs used in the realm of energy production (geothermal, CCS, gas-storage or spent-radionuclide repositories) contain a number of solid and fluid phases, the latter being found in mobile- and immobile-fluid regions. Performance and lifetime of a particular georeservoir depend on the volumes and/or interface areas of some of these regions and/or phases. Mostly, these cannot be measured by geophysical and hydraulic methods. Since they essentially relate to fluid-based transport processes, attempting to measure them by means of tracer tests is a reasonable endeavor (Ghergut et al. 2013a). In the sequel, we examine the possibility to quantify fluid and solute transfer between georeservoir compartments by means of artificial tracers undergoing first-order kinetic exchange processes (alongside with reference tracer species that do not partition between georeservoir compartments).

In order to evaluate the performance of tracer tests, we resort to two different kinds of modeling. We first set up a conceptual model (governing equations for transport processes, and their approximation), then we define five different transport scenarios of first-order kinetic exchange, which we consider to roughly capture the broad variety of exchange processes of interest (matrix diffusion, partitioning and in particular adsorption-desorption). For each of these transport process scenarios, we use a finite-element (FE) model to simulate tracer signals ('breakthrough curves', BTC) from inter-well tests. Finally, as an additional consistency check, we derive the apparent flow-storage repartition (FSR) functions equivalent to the FE-simulated BTCs, and find that these respond to transport parameter variations in a consistent manner. Whereas the FE approach belongs to the distributed-parameter modeling, the FSR approach is essentially a non-parametric one.

2. CONCEPTUAL MODEL, AND GOVERNING EQUATIONS FOR THE FE SIMULATIONS OF TRACER TRANSPORT

The advective-dispersive transport of a tracer species subject to exchange processes between mobile-fluid and either solid or immobile-fluid regions (but else physico-chemically stable) is governed by the partial differential equation:

$$\phi_f \frac{\partial C_f}{\partial t} = \underbrace{\nabla \cdot (D_f \cdot \nabla C_f)}_{dispersion} - \underbrace{\mathbf{q} \cdot \nabla C_f}_{advection} - \underbrace{F_m}_{exchange} \quad (\text{cf. Nomenclature section at the end of the paper})$$

If the process underlying the F_m term is ‘matrix diffusion’ (see Carrera et al. 1998 for details), then F_m can be written either in terms of a concentration gradient corresponding to Fick’s first-order law of diffusion, as:

$$F_m = \sigma_m \phi'_m D_m \left. \frac{\partial C_m}{\partial \zeta} \right|_{\zeta=0}$$

or, by virtue of mass balance over finite-size matrix blocks, as:

$$F_m = -\sigma_m \phi'_m D_m \frac{\partial C_{m,AVE}}{\partial t}$$

For C_m , the diffusion equation holds:

$$\sigma_m(\zeta) \frac{\partial C_m}{\partial t} = \frac{\partial}{\partial \zeta} \left(\sigma_m(\zeta) D_m \frac{\partial C_m}{\partial \zeta} \right)$$

subject to initial and boundary conditions:

$$C_m(x, \zeta, t=0) = 0$$

$$C_m(x, \zeta=0, t) = C_f(x, t) \text{ at the matrix-fracture interface (where } \zeta=0)$$

$$\left. \frac{\partial C_m}{\partial \zeta} \right|_{(x, \zeta = \zeta_{\max}, t)} = 0$$

(to be noted, the maximum depth within matrix blocks ζ_{\max} is related to the effective matrix block size L_m by a characteristic relationship depending on the geometry of matrix blocks – see Carrera et al. 1998 for details)

If the processes underlying the F_m term are kinetic partitioning processes (in particular, when the immobile-fluid region is within the solid rock body, such processes are usually deemed ‘adsorption – desorption’), then F_m can be written, in first-order approximation,

$$F_m = \phi_m (-\alpha C_f + \beta C_m)$$

whereas in the immobile-region C_m follows

$$\frac{\partial C_m}{\partial t} = \alpha C_f - \beta C_m$$

Further, according to Haggerty and Gorelick (1995), Carrera et al. (1998), Haggerty et al. (2001), this linear expression of kinetic exchange processes can (under certain circumstances) also be applied to approximate ‘matrix diffusion’ processes, by setting

$$\alpha \equiv \beta \equiv \frac{D_m}{L_m^2}$$

with L_m as defined previously. To be noted, α and β have the physical dimensions of 1/time (like a normalized reaction rate, or a ‘turnover’ rate). Since the most natural choice of a time scale is the fluid residence time in the mobile-fluid compartment (MRT , total pore volume in this reservoir compartment, divided by the prescribed flow rate), we shall use MRT^{-1} to scale the exchange rates.

Closed-form solutions to the above equation system can be found only for very simple flow-field geometries, such as parallel or radial monopole flow (Maloszewski and Zuber 1985, 1993). In many cases of practical interest, these equations can be solved only by numerical methods. This can be done, for instance, by using the commercially available software FEFLOW developed by Diersch (2014). For the purposes of this study, we set up a porous mono-continuum model (without fracture elements), and implement the above equations by means of the ‘multi-reaction’ option, with rate coefficients α and β multiplied by the porosity of each compartment, consistently with FEFLOW’s internal representation of solute reaction terms.

3. MODEL DOMAIN AND PARAMETRIZATION, TRANSPORT SCENARIOS, AND FE SIMULATION RESULTS

The FE model describes the radially-divergent monopole injection of fluid and tracer into a homogeneous porous layer ('aquifer') of total effective aperture (porosity \times thickness) of 0.15 m, and hydraulic transmissivity (conductivity \times thickness) of 3.5×10^{-3} m²/d, under a fluid injection rate of 100 m³/d. This rate value requires a radial length of at least 80 m, for the tracer plume to still stay well within the model domain (far enough from its 'remote boundary') at the end of the simulated tracer test duration of 20 days (cf. visualizations of the tracer's late plume at the end of this section). By virtue of cylindrical symmetry, the model domain can be reduced to 1-D.

Tracer is added as a short pulse (a well-defined total mass of tracer, added over a time interval of about 0.01 day; pulse shape is unimportant). Tracer signals ('breakthrough curves', BTCs) are recorded in flux mode at two passive observation points (no forced-gradient sampling!) in 30 m and 50 m distance from the injection boundary (cf. visualizations at the end of this section). Observing the Courant and Peclet number criteria, spatial discretization is by up to 1500 elements, of refined element size in the vicinity of the injection boundary, with maximum element length not exceeding half of the dispersivity value; time discretization uses variable (adaptive) time steps not exceeding 0.02 d, and much shorter time steps (10^{-10} d) at early times.

We choose to present five solute transport scenarios (table 1) that are meant to summarize the most typical situations of interest in georeservoir field testing practice. For a simulated tracer test duration of 20 days, each scenario simulation takes less than 50 seconds to complete on a 3.7 GHz machine with 2.2 GB of effectively-available RAM (running under Windows XP). The simulated BTCs are shown in fig. 1, in both linear and logarithmic scaling, to enable a good picture of peak intervals, and, respectively, of tailing details.

Table 1: Generic exchange model parametrization

Notation	α value [MRT^{-1}]	β value [MRT^{-1}]	approx. fixation : release ratio	fluid-rock interface density	physical interpretation
f1_s1	0.15	0.15	1 : 1	(medium)	matrix diffusion or kinetic partitioning
f1_s03	0.15	0.05	1 : 1/3	(medium)	kinetic exchange (fluid-fluid partitioning or adsorption-desorption)
f1_s01	0.15	0.015	1 : 1/10	(medium)	kinetic exchange (fluid-fluid partitioning or adsorption-desorption)
f3_s3	0.45	0.45	3 : 3	(high)	matrix diffusion or kinetic partitioning
f03_s03	0.045	0.045	1/3 : 1/3	(low)	matrix diffusion or kinetic partitioning

From fig. 1, it can be seen that:

- peak height is roughly determined by the fixation rate α ; peak height decreases with increasing fixation rate;
- peak arrival times and peak interval durations (before the onset of late tailings) depend on both α and β ;
- the tailing decrease rate is roughly determined by the release rate β ; and it gets accelerated with increasing β ;
- the higher the release rate β , the lower the height of late tailings;
- the lower the release rate β , the sooner the onset of late tailings for a given fixation rate α ;
- for matrix diffusion processes in first-order approximation, the higher the fluid-rock interface area density, the later and lower the peak value, the higher and faster the mid-term tailings, but the lower and longer the late tailings.

This is largely consistent with expectations, and tracer signal changes exhibit sufficient sensitivity to enable parameter determination, at least in principle (with further aspects to it as discussed in the next section). Figure 2 enables to compare the radial extent of tracer spreading between the different scenarios; the radial distribution of tracer concentrations in the mobile-fluid compartment at the end of the experiment is displayed with the labels $f\#_s\#$, and accordingly for the immobile-fluid or rock compartments, with the labels $s\#_f\#$.

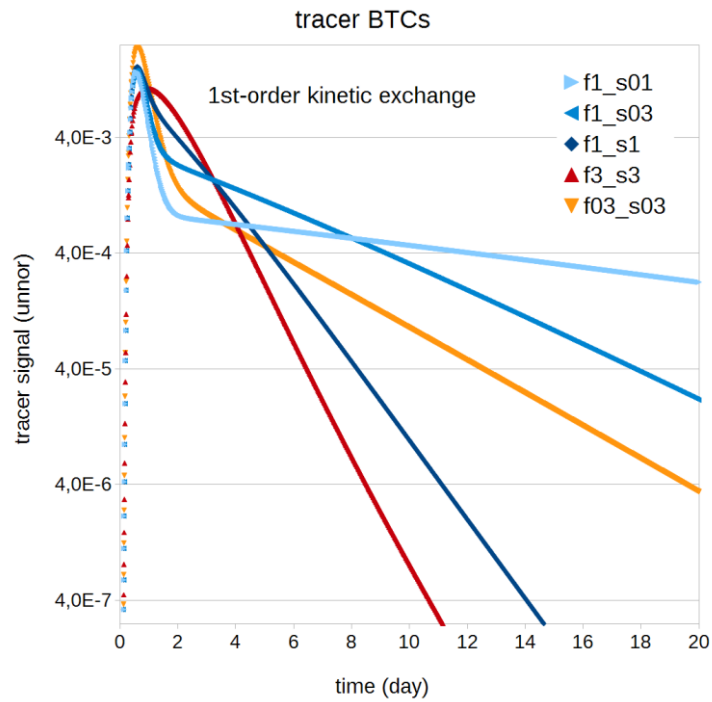
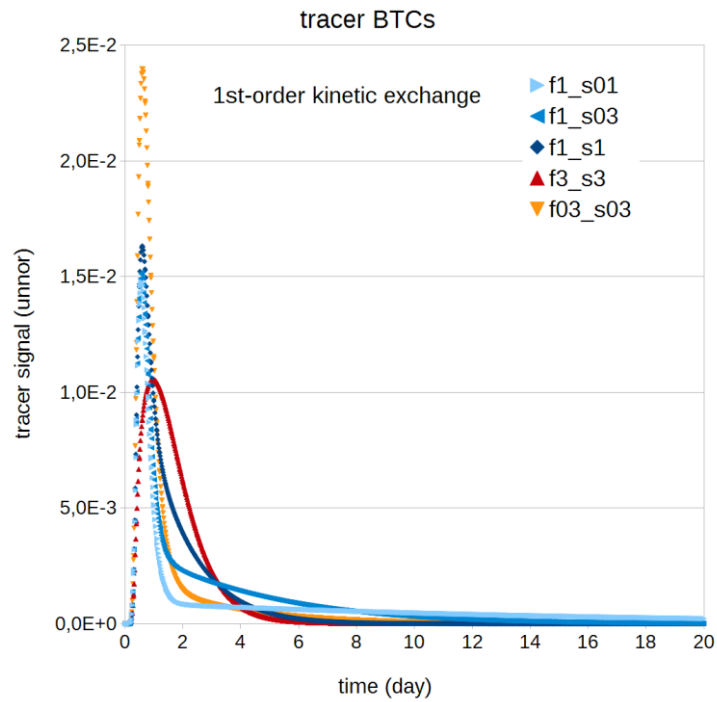


Figure 1: Tracer breakthrough curves (BTCs) obtained by FE model simulations; upper section: linear scaling of tracer concentrations (focus on BTC peak heights); lower section: logarithmic scaling of ibid. ('zooming in' into BTC tailings).

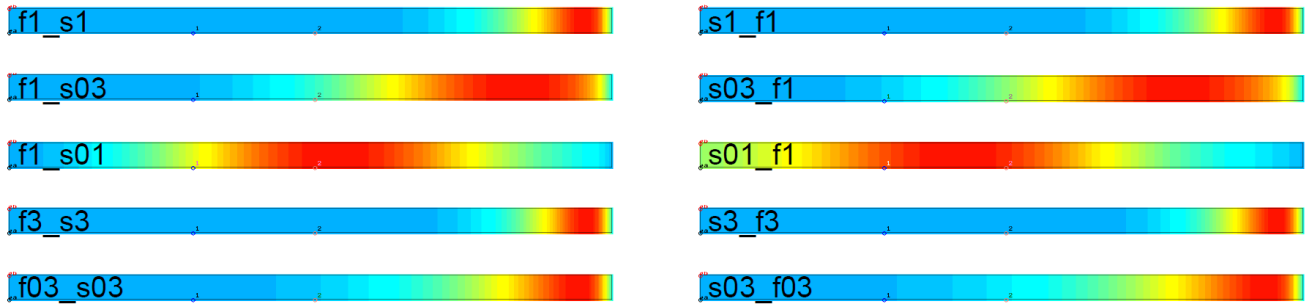


Figure 2: Radial extent of tracer spreading at the end of the test (20 days after injection), shown for each of the five transport scenarios, within the mobile-fluid compartment (l.-h.s.) and within the rock or immobile-fluid region (r.-h.s.).

4. CONSISTENCY CHECK IN APPARENT-FSR TERMS, INTERPRETATION AND DISCUSSION

In Behrens et al. (2010), Ghergut et al. (2007, 2013^b), it had been recommended to use flow-storage repartition (FSR) shapes (originally introduced by Shook 2001, for characterizing what M.G. Shook deemed as ‘fractured-reservoir geometry’) also for the broader purposes of dual-porosity, or fractured-porous media characterization in the presence of kinetic exchange processes between mobile- and stagnant-fluid compartments (whereas Shook’s original analysis only considered advective-dispersive processes). FSR analysis is a versatile tool for characterizing subsurface flow and transport systems.

FSR can be derived from tracer signals measured in inter-well tests, if certain requirements (Ghergut et al. 2013^b) are met – basically, the same as required for equivalence between the fluid residence time distribution (RTD) and the measured inter-well signal (pre-processed and de-convolved if necessary, as described by Shook 2001) of a conservative tracer. In a more general approach (Ghergut et al. 2007), a FSR is derived from a RTD as a trajectory in normalized $\{1^{\text{st}}, 0^{\text{th}}\}$ -order statistical moment space; more intuitively, as a parametric plot of 0^{th} -order against 1^{st} -order statistical moments of RTD truncated at time t , with t as a parameter running from the first tracer input to the latest available tracer sampling; with 0^{th} -order moments being normalized by the total tracer recovery, and 1^{st} -order moments by the mean RT. Fracture-dominated systems plot in the upper left (high F, low S) region of FSR diagrams; ‘plug’ flow in a homogeneous, dispersion-less mono-continuum (Peclet number $Pe = \infty$) displays as a straight line from $\{F, S\} = \{0, 0\}$ to $\{F, S\} = \{1, 1\}$. This analysis tool appears particularly suited for characterizing porous-fissured formations like those targeted by geothermal exploration in the South-German Malm-Molassebecken.

Figure 3 shows the apparent-FSR shapes derived from the tracer BTCs (fig. 1) that have been obtained by FE simulations as explained and discussed in the previous section. From fig. 3, it can be seen that:

- for a given α , the flow capacity of any given storage capacity decreases with increasing β (which seems surprising at first sight); in other words, with decreasing imbalance between the fixation rate α and the release rate $\beta \leq \alpha$, the apparent FSR shape approaches the diagonal shape of a uniform ‘plug-flow’ system;
- for a given release rate β , increasing the fixation rate $\alpha \geq \beta$ will reduce the flow capacity at high storage values, while raising the flow capacity at low storage values;
- for matrix diffusion processes in first-order approximation, the higher the fluid-rock interface area density, the lower the flow capacity for any given storage capacity;
- for matrix diffusion processes in first-order approximation, the higher the fluid-rock interface area density, the higher the storage capacity for any given flow capacity;
- for matrix diffusion processes in first-order approximation, the higher the fluid-rock interface area density, the closer the apparent-FSR shape will approach the diagonal; this looks surprising at first sight, but can be understood from the fact that, when $\alpha = \beta \gg \gg MRT^{-1}$, the matrix diffusion process approaches an equilibrium-retardation process, to which the apparent FSR becomes insensitive, while on the other hand the first-order approximation suggested by Haggerty and Gorelick (1995), Carrera et al. (1998), Haggerty et al. (2001) becomes increasingly inadequate for describing matrix-diffusion processes.

Some of these findings are somewhat surprising at first sight, but become understandable by comparing the degree of imbalance between fixation (α) and release (β) with the degree of deviation from the diagonal shape of the uniform ‘plug-flow’ system.

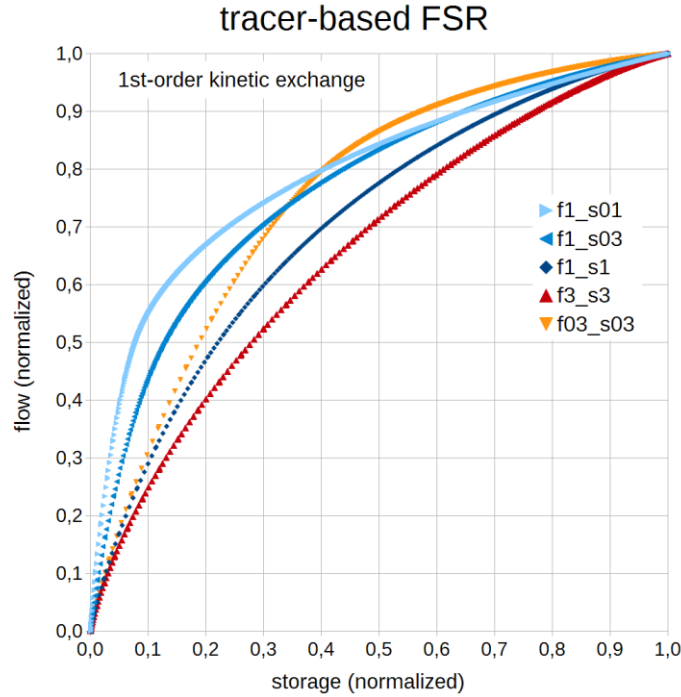


Figure 3: Apparent FSR shapes derived from simulated tracer BTCs for five typical transport scenarios.

5. CONCLUDING REMARKS

Tracer tests are seen to be a suitable tool for evaluating kinetic exchange processes in dual-porosity media, in particular matrix diffusion processes in fissured or fractured formations.

Simulated tracer BTCs (fig. 1) for five typical kinetic-exchange process scenarios (roughly covering the broad range of solute partitioning processes that may be relevant for georeservoir characterization) are seen to respond to variations of the kinetic-exchange parameters in a sensible, and fairly-sensitive manner. Apparent flow-storage repartition shapes (fig. 3) derived from simulated tracer BTCs are seen, as well, to consistently reflect variations in kinetic-exchange parameters.

However, in real-world applications, in order to be able to conduct ‘parameter inversion’ from measured tracer BTCs, e. g., for estimating fluid-rock interface densities, it would be necessary to have diffusive or sorptive tracer species with well-defined adsorption/desorption rates and/or reliably-known diffusion coefficients. These, in turn, depend on the in-situ physicochemical conditions (temperature, pH, ‘salinity’ or ionic strength). They need to be determined by accompanying laboratory experiments, independently of the field application, however using fluid and rock material that is representative of the target georeservoirs. This is a sometimes feasible, but not trivial task.

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7. NOMENCLATURE

Notation and symbols used in section 2, largely following Carrera et al. (1998), are listed and briefly commented below.

ϕ_f	porosity of the mobile-fluid region
D_f	dispersion coefficient tensor for the mobile-fluid region

q	flow rate
C_f	solute concentration in the mobile-fluid region
F_m	solute flux term representing exchange processes between mobile- and immobile-fluid regions
ϕ'_m	intrinsic porosity of the rock matrix, related to the immobile-fluid region (bulk) porosity ϕ_m by $\phi_m = (1 - \phi_f) \times \phi'_m$
D_m	molecular diffusion coefficient within the rock matrix pore space
$\sigma_m(\zeta)$	matrix rock surface area per bulk aquifer volume, at depth ζ within a matrix block
σ_m	abbreviates $\sigma_m(\zeta) _{\zeta=0}$ and can also be expressed in terms of an effective, finite matrix block size L_m , by virtue of the rock volume balance: $\sigma_m L_m = 1 - \phi_f$
C_m	solute concentration within the matrix rock pore space (immobile-fluid region)
$C_{m,AVE}$	matrix-block-averaged solute concentration within the immobile-fluid region

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