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Peak and tail scaling of breakthrough curves in hydrologic tracer tests

T. Aquino *, A. Aubeneau, D. Bolster

Department of Civil & Environmental Engineering & Earth Sciences, University of Notre Dame, 46556 IN, USA

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ABSTRACT

Power law tails, commonly observed in solute breakthrough curves, are notoriously difficult to measure with confidence as they typically occur at low concentrations. This leads us to ask if other signatures of anomalous transport can be sought. We develop a general stochastic transport framework and derive an asymptotic relation between the tail scaling of a breakthrough curve for a conservative tracer at a fixed downstream position and the scaling of the peak concentration of breakthrough curves as a function of downstream position, demonstrating that they provide equivalent information. We then quantify the relevant spatiotemporal scales for the emergence of this asymptotic regime, where the relationship holds, and validate our results in the context of a very simple model that represents transport in an idealized river.

1. Introduction

Rivers are the backbone of environmental flows. Distilled rain water acquires dissolved solutes and suspended particulates as it travels on hill-slopes. This water discharges into the river network, where it travels over considerable distances as rivers link landscapes over continental scales [31]. Rivers also act as filters by processing and transforming the load they carry, influencing the biogeochemistry of downstream water bodies [35]. Thus, understanding the processes responsible for the physical translocation and the biogeochemical transformations of upstream inputs to downstream outputs is critical to scientists, stakeholders and decision makers.

Streams and rivers are complex, heterogeneous systems, with fast surface flow transporting substances quickly in the main channel and slow boundary layer and subsurface flow retaining substances for potentially long periods of time. This broad separation of velocities and associated time scales leads to anomalous transport, which cannot be adequately described with traditional one-dimensional Fickian advection dispersion models [33]. The trapping of solutes in a river's bed-sediment leads to heavy-tailed residence times which manifest as power law tails in experimental breakthrough curves (BTCs) [25]. These heavy tailed BTCs demonstrate long-term retention of solutes in rivers, which is particularly important for the many biogeochemical processes that occur in the slow regions near or inside the river-bed [6,16,26,34].

In traditional tracer tests, a pulse (or drip) of tracer is released and its concentration over time is measured at some downstream location(s) to obtain BTCs. The mass of stream-borne dissolved solutes entering the bed is often only a small fraction of the total mass and it is further diluted upon return to the open channel. The signals associated with tracers that have traveled through the bed therefore appear at very low concentrations in measured BTCs, often orders of magnitude below peak concentrations [21]. This poses a significant experimental challenge as reliable and sufficiently sensitive measurements can be difficult to obtain. Typical methods based on electric conductivity resolve only 2 to 3 orders of magnitude, while fluorescent dies can resolve over 4 orders of magnitude. Even though isotopic tracers can be very sensitive, up to 6 orders of magnitude for stable isotopes and 8 or 9 for radioactive tracers, they are seldom used and most experiments only resolve relatively short timescales [14,40]. Conversely, the peak concentration of a BTC from a pulse injection is a reliable measurement, because it is typically much larger in magnitude. The change in peak concentration with downstream distance could therefore provide reliable evidence of anomalous transport characteristics.

Any apparent mass loss in the BTC of a conservative tracer must have been retained during transport and should eventually leak back to the main flow. This would be true for example in flumes, or rivers on bedrock without connection to a regional aquifer. Even when there are gains and losses through groundwater exchange, this concept remains valid so long as a proper mass balance is enforced [17]. The mass lost from the BTC compared to the mass actually injected upstream (ignoring the flowpaths bypassing the sampling location) should thus reappear as a tail if the instruments have sufficient sensitivity and sampling occurs over sufficiently long times. We argue that a dynamical relationship therefore exists between the bulk of the solute, which is transported directly by the water column in the river, and the solute mass that reenters the

* Corresponding author.
E-mail address: tdecampo@nd.edu (T. Aquino).

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water column after being retained in the sediment bed. If we consider a BTC measured at a fixed location, the fraction of solute that has spent a considerable time in the sediment bed will define the long-time tailing behavior. Since the mass recovered in the tail was "lost" from the main channel flow, one should expect an equivalent signal missing from the main pulse, which in turn should also appear as a faster than expected decay in the peak of the BTC downstream [4]. Thus, if we consider multiple BTCs measured at different downstream positions, we may ask the following question: Given the behavior of the peak value of the BTCs at multiple positions, can we infer the tailing properties of a single BTC at a fixed position? This idea is illustrated in Fig. 1. A relationship of this type would allow one to infer details about the solute transport occurring in the sediment bed from measurements of the bulk mass transported in the water column. This would provide an alternative method to assess the behavior of BTC tails through measurements of peak concentrations.

The present work is structured as follows. Section 2 presents our model: we derive a general late-time relation between the scaling of the tail of a BTC for a conservative tracer at a fixed downstream position and the scaling of the BTC peak as a function of downstream position. Section 3 is dedicated to illustrating the general results of Section 2 in a concrete scenario, so as to clarify the roles of the underlying physical processes. For this purpose, in Section 3.1 we construct a very simple conceptual model for river transport, which is deliberately chosen to be simple, yet complex enough to demonstrate the desired behaviors. We then determine the relevant spatiotemporal scales for the onset of the asymptotic scaling behavior in the context of this model, and also discuss the pre-asymptotic regime. We validate our results using numerical particle tracking simulations in Section 3.2. An overall discussion is presented in Section 4.

2. Asymptotic behavior of tail and peak scaling

In order to address the question of the relationship between peak and tail scaling in BTCs, it is necessary to describe solute transport in a sufficiently general framework that allows the relationship between retention in the sediment bed and transport in the water column to come to light. Particle-based random walk methods, whether from a theoretical or numerical (particle tracking) perspective, have been used extensively to represent solute transport in flows across a diverse range of hydrologically relevant flows [27,37]. In particular, the related idea of subordination has been used to derive results on peak, tail and moment properties of BTCs for solute transport in heterogeneous porous media [5]. The basic premise of particle-based methods is to discretize the solute plume into a discrete number of individual particles, each of which then move based on probabilistic rules that aim to capture microscopic and macroscopic processes of the system of interest in an effective manner. It is important to note that these pseudo-particles are abstract theoretical (or numerical) devices and do not aim to represent actual individual solute particles [10,15]. They are characterized by effective properties that depend on the specific solute, flow and background medium. These particles are tracked as they move due to advection by the background flow and dispersion according to an appropriate stochastic process representing the dispersive properties of the solute in a particular medium. From a numerical standpoint, particle tracking methods have the benefit of being essentially free of numerical dispersion [13]. From both the numerical and theoretical point of view, these methods provide a very flexible framework to represent transport phenomena, ranging from classical (Gaussian) advective–dispersive transport [28] to more general processes [8,9]. The present work builds on this type of approach to explore peak and tail scaling properties in the context of river and stream transport. Importantly, these methods take into account stochastic properties in a natural fashion. Furthermore, they allow us to derive our results without the necessity of imposing overly restrictive assumptions on the nature of the transport, and they can thus be applied to a variety of natural systems.

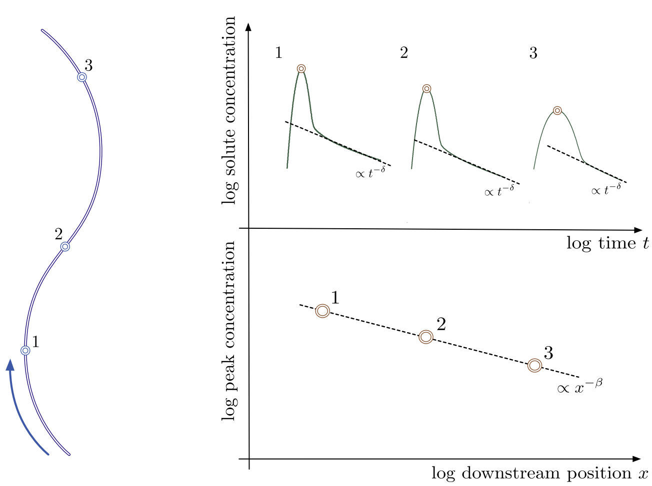

Fig. 1. Given the behavior of the peak value of the BTCs measured at multiple downstream positions (decay exponent β), can we infer the tailing properties in time of a single BTC at a fixed position (decay exponent δ)? Given a number of stations positioned at different downstream positions along a river or stream (left panel), the temporal tail scaling at each position is expected to follow some power law scaling $t^{-\delta}$ at late times (top right panel). We ask the question of whether this behavior has a discernible signature in the peak decay as a function of downstream position, where we expect some asymptotic power law scaling $x^{-\beta}$ (bottom right panel).

2.1. Theoretical framework

In classical random walk approaches, time is discretized and all particles move over a fixed time step according to specific stochastic transport process (e.g. Brownian motion [36], Fickian dispersion [37]). For our theoretical description, we adopt an alternative view often called the continuous time random walk [9] whereby we fix a specified spatial distance along the downstream dimension rather than fixing the time step. We then ask: what is the probability that a particle takes a certain amount of time to traverse this fixed length? In this description, the randomness in the movement of the particles is encoded in the density of waiting times needed to traverse this fixed length. For example, when modeling river transport, a particle that travels through the water column will take a much shorter time to travel a fixed distance than one that is retained in the sediment bed and later released back into the main channel. Similar approaches, where a random process is modified by some waiting time distribution that characterizes an inactive or immobile phase, can be found in [5,8], which rely more explicitly on the related concept of subordination. An overview of the related approaches of fractional advection–dispersion, subordination and continuous time random walks can be found in [33]. To our knowledge, the approach presented here is new in the context of river and stream solute transport and provides a clear picture of the physical processes and assumptions involved.

Let us formalize our ideas. We wish to describe the motion of a particle of solute undertaking random motion starting from a known position x_0 at time t_0. Let x be position downstream, and start by fixing a length l. Assuming that we are interested in length scales over which the movement of a particle is independent of previous history, we can describe the motion of our particle by: