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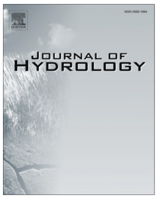

Assessment of transient storage exchange and advection–dispersion mechanisms from concentration signatures along breakthrough curves

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SUMMARY

Solute transport in rivers is controlled by surface flow hydrodynamics and by transient storage in dead zones, pockets of vegetation and hyporheic sediments where mass exchange and retention are governed by complex mechanisms. The physics of these processes are generally investigated by optimization of transient storage models (TSMs) to experimental data often yielding inconsistent and equifinal parameter sets. Uncertainty on parameters estimation is found to depend not only on the rates of exchange between the stream and storage zones, the stream-water velocity and the stream reach length according to the experimental Damkohler number (DaI), but also on the relative significance between transient storage and longitudinal dispersion on breakthrough curves (BTCs). An optimization strategy was developed and applied to an experimental dataset obtained from tracer tests in a small lowland river, analyzing BTCs generated through tracer injections under different conditions. The method supplies a tool to estimate model parameters from observed data through the analysis of the relative parameter significance. To analyze model performance a double compartment TSM was optimized by a regular fit procedure based on simple root mean square error minimization and by a fit based on a relative significance analysis of mechanism signatures. As a result consistent longitudinal dispersion and transient storage parameters were obtained when the signature targeted optimization was used.

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1. Introduction

The characterization of surface dispersion induced by velocity gradients in a turbulent flow and its distinction from transient storage is one of the major problems of solute transport models application. Evaluation of dispersion parameters was firstly investigated by Taylor (1954) and then by Fischer et al. (1979) by estimation of a longitudinal dispersion coefficient from shear velocity profiles. This approach resulted to be unreliable to predict transport in natural rivers (Nordin and Troutman, 1980) where transport mechanisms are complex, and non-Fickian mixing processes concur in increasing the skewness of breakthrough curves (BTCs). Chatwin (1971) proposed a simplified method to determine the longitudinal dispersion coefficient from experimental datasets, suggesting that the dispersion coefficient can be estimated from the leading edge of a BTC produced by a slug injection of tracer. Many other attempts to predict the dispersion coefficient were made (Liu, 1977; Deng et al., 2001; Seo and Cheong, 1998; Koussis and Rodríguez-Mirasol, 1998), but all the empirical, semi-empirical and theoretical predictions of longitudinal dispersion apply only to specific river systems. In fact, it is a shared opinion that the dispersion coefficient depends on too many factors and that its prediction is always an arduous task.

Different mathematical models have been formulated to simulate and analyze surface transport in rivers coupled with exchange with storage areas and hyporheic zones. According to the Transient Storage Model (TSM) (Thackston and Schnelle, 1970; Bencala and Walters, 1983), the mass exchange between the main channel and the storage zones can be represented as a first order mass transfer mechanism. The TSM was later applied in a simplified version by Davis and Atkinson (2000), who developed an Aggregated Dead Zone model (ADZ). The ADZ divides the channel cross section in two parallel regions: the bulk flow and the storage area, where the latter is accounting for both storage and dispersion, whereas the flow area accounts only for longitudinal advection. An extended version of the TSM was presented by Choi et al. (2000), who proposed a TSM with multiple storage zones. Wörman et al. (2002) developed the Advective Storage Path (ASP) based on Elliott and Brooks (1997) work on bedform induced hyporheic exchange. A Continuous Time Random Walk model (CTRW) was proposed by Boano et al. (2007), whereas Deng et al. (2006)

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suggested a fractional dispersion model. Haggerty et al. (2000) proposed an advection–dispersion mass transfer equation in which the transient storage is expressed through a convolution integral of the in-stream concentration and an exponential residence time distribution and developed a Multi Rate Mass Transfer model (MRMT) (Haggerty and Gorelick, 1995; Haggerty, 2002). A comprehensive review of the models developed in the last decades is presented by Boano et al. (2014).

Davis and Atkinson (2000) found that after a transitional phase the transient storage mechanism dominates the shear flow dispersion (i.e. the dispersion induced by velocity gradients). Application of the ADZ demonstrated that longitudinal dispersion can be well represented when considered as a bulk effect of only transient storage processes. Choi et al. (2000) found that in most cases a single compartment storage model can properly fit concentration data, but how parameters represent the associated processes was not investigated. Bottacin-Busolin et al. (2011) observed that transient storage leaves signature at different timescales, suggesting that a double compartment model can better represent the exchange with the storage zones.

Wagner and Harvey (1997) used for the first time a sensitivity analysis to investigate how concentration model output depends on each parameter, concluding that parameters identifiability depends on the Damkohler number (Bahr and Rubin, 1987). Gooseff et al. (2013) used sensitivity analysis to investigate how parameters of a single compartment TSM depends on reach lengths: in their work parameter optimization and sensitivity analysis were performed using UCODE (Poeter and Hill, 1998), a numerical code universally applicable for reverse modeling. Kelleher et al. (2013) used the global sensitivity indices for nonlinear mathematical models (Sobol, 2001) to investigate how different stream morphologies influence the identifiability of storage and transport parameters. In the two works cited above the sensitivity analysis was applied after parameter evaluation to evaluate if a TSM can consistently reproduce transient storage dynamics.

The main idea of this work is to provide a tool to improve the reliability of estimated parameters. For this purpose, we extend sensitivity to a relative significance analysis between parameters, in order to identify BTC segments carrying clear signatures of either dispersion or transient storage. We applied a double compartment TSM to an experimental dataset from a natural stream, obtained by sampling BTCs after plateau injections of a fluorescent tracer under different conditions. Once the relative significance is analyzed, dispersion and storage parameters are optimized over BTC segments where the respective mechanism signatures are dominant.

2. Experimental material and methods

2.1. Description of the experimental work

The lowland stream Erpe is a small spring river located at the eastern edge of Berlin. The Erpe is polluted by intense agriculture in its catchment and by the discharge from various wastewater treatment plants located along its course (Horner et al., 2009). Due to the high level of pollution, the Erpe was chosen as a case study for other experimental works focused on the effects of nutrients and pollutants on the stream (Gücker and Pusch, 2006; Gücker et al., 2006) and on the hyporheic zone (Lewandowski et al., 2011). In these works the river was investigated analyzing stream water and sediments samples, using different techniques such as temperature-based measurements of the stream-sediments exchange or water tracing with sodium chloride. In the present study we used a fluorescent dye (Rhodamine WT) to trace water transport and storage mechanisms. Experiments were conducted in two field campaigns, the first performed in October 2012 and the second in September 2013.

The test reach was divided in three segments, bordered at the upstream and downstream ends by four stations numbered from 0 to 3. Station 0 was located at 52°29′32.1″N and 13°39′01.2″E, station 1 at 52°29′ 26.2″N and 13°38′59.0″E, station 2 at 52°29′22.7″N and 13°38′56.1″E, station 3 at 52°29′17.8″N and 13°38′49.0″E. For each reach we roughly estimated the stream width and the stream depth during both campaigns to measure the flow area, as reported in Table 1. The studied reach is 650 m long and about 3 m wide, with several bends but no distinct meanders (see Fig. 1). The flow sections are homogeneous along the longitudinal distance for each reach and the ratios between mean depth and mean width is $d_W/b_W = 0.15$. Bed sediments consist mainly of organic-rich silt with submerged vegetation present along the entire river course. The stream is characterized by submerged macrophytes growing from May to September and removed by the local water management authorities in autumn. During field works riverbanks were covered with tall grass, except in 2013 when submerged and bank vegetation was more developed than in 2012.

The distance between stations was chosen accounting for complete transverse mixing, according to Fischer et al. (1979):

$$L = 0.1\frac{Ub_W^2}{\epsilon_t} \tag{1}$$

where U the mean flow velocity (m s^{-1}), L the reach length (m) and ϵ_t is the transverse mixing coefficient ($\text{m}^2\ \text{s}^{-1}$), given by the following relation, valid for slightly irregular channels:

$$\epsilon_t = 0.40 d_W u^* \tag{2}$$

where d_W is the flow depth (m) and u^*, assuming uniform flow, is the shear velocity (m s^{-1}). The length for full transverse mixing, evaluated from Eq. (1), ranged from $L = 50$ to $L = 60$ m for the two field campaigns, so the chosen reach lengths fulfilled transverse mixing requirements (see Table 1).

Flow rates and water levels were constant during experiments. Water discharge, measured with a propeller flow meter, was constantly $Q = 0.362\ \text{m}^3\ \text{s}^{-1}$ and $Q = 0.190\ \text{m}^3\ \text{s}^{-1}$ during the 2012 and the 2013 campaigns respectively.

Three submersible fluorometers for Rhodamine WT detection were used: a SCUFA submersible fluorometer, with minimum detection limit of 0.04 ppb, and two YSI 6920 equipped with an optical probe for Rhodamine WT with minimum detection limit of 0.1 ppb. Fluorometers where calibrated using nine standard Rhodamine WT solutions ranging from 0 to 80 ppb. An additional Albillia GGUN-FL30 field fluorometer was used at one reach during the second campaign.

Four experiments were conducted: three were performed in October 2012, whereas the fourth experiment was conducted in September 2013. Experiments differed in reach lengths, injection duration, and water discharge (see Tables 1 and 2). The tracer was injected with a peristaltic pump to generate a constant concentration at station 0. Peak concentration ranged from 39 to 63 ppb and the concentrated solution was injected at a rate of 0.91 ml/s. Fluorometers were located in the mid of the channel at

Table 1
Subdivision of the Erpe stream in sub-reaches. Each reach is composed by the abbreviated river name and by a number referencing to the upstream and the downstream station. Water depth is reported for both the experimental campaigns conducted in 2012 and 2013, with symbols $d_{W,2012}$ and $d_{W,2012}$ respectively.

Reach ID	L (m)	b_W(m)	$d_{W,2012}$ (m)	$d_{W,2013}$ (m)
Er-01	162.2	3.5	0.45	0.50
Er-12	162.7	3.0	0.45	0.50
Er-23	195.9	3.5	0.45	0.50

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