# A Comparison of Three Solute Transport Models Using Mountain Stream Tracer Experiments

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Abstract Stream ecology may be influenced by the temporary trapping of solutes in geomorphologic structures, which is usually quantified by fitting the Transient Storage Model to tracer data. This paper explores the relationships between the parameters of this model and those of two simpler models, namely the Advection-Dispersion Model and the Aggregated Dead Zone model. It is motivated by the possibility of obtaining more reliable transient storage parameter values by correlating them with the parameters of the other models instead of evaluating them directly. Results were obtained by fitting all three models to a set of tracer data from mountain streams, predominantly in Iceland. Some strong correlations were found between some of the parameters of the transient storage model and the advection-dispersion model, but no strong correlations were found between the parameters of the transient storage model and the aggregated dead zone model. For all three models, combinations of the optimized parameters correctly described the bulk movement of the solute cloud, giving confidence in the optimized parameters.

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#### 1 Introduction

In this paper we compare three models of solute transport for mountain streams. We examine the models' effectiveness at describing the transport of a tracer through many stream reaches and, by fitting them to tracer data, elucidate meaningful parameters that describe the streams physically. The work is part of a larger study investigating the effect of temperature and water transient storage on stream metabolism and nutrient cycling (Friberg et al. 2009; Woodward et al. 2010; Demars et al. 2011a, b; Gudmundsdottir et al. 2011; Manson et al. 2011; Rasmussen et al. 2011; O'Gorman et al. 2012; Hannesdottir et al. 2013). Information from 66 tracer experiments executed in several geothermal areas in Iceland (Hengill, Hveragerdi, Hveravellir, Kerlingarfjöll, Torfajökull and Vonarskard) was combined into a single data-set. Four additional experiments from similar geothermal and climatic sites in Kamchatka, Russia, were also used (O'Gorman et al. 2014).

In much previous work the so-called Transient Storage Model (e.g. Bencala and Walters 1983) has been fitted to tracer data in order to evaluate the parameters that quantify the transient storage process. Although good fits to the data are often achieved, questions have been raised over how well the parameters can be identified (e.g. Wagner and Harvey 1997; Wagener et al. 2002; Worman and Wachniew 2007). Here we are motivated by the possibility of obtaining the parameters from correlations with the parameters of alternative models. Hence we seek to explore relationships between the parameters of the Transient Storage Model, the Advection Dispersion Model and the Aggregated Dead Zone Model.

## 2 Solute Transport Models

This section provides background information on the three models used, namely the Advection-Dispersion Model, the Transient Storage Model and the Aggregated Dead Zone Model.

## 2.1 The Advection-Dispersion Model (ADM)

In the ADM, advection refers to the movement of a solute caused by the longitudinal velocity of flow and dispersion refers to the longitudinal spreading of solute that takes place simultaneously with the advection. The advection is characterised by the cross-sectional average longitudinal flow velocity. In reality, solute is carried faster than the average velocity in the deeper parts of a river cross-section and is carried slower than the average velocity in the shallower parts of a river cross-section. The longitudinal spreading is caused by the interaction between this cross-sectional variation in longitudinal advection and mechanisms causing

cross-sectional mixing, which also vary throughout the cross-section. In the model, turbulent diffusion and secondary currents are responsible for the cross-sectional mixing. The overall spreading effect is quantified by the dispersion coefficient and the process is often termed shear flow dispersion. Once a solute cloud has been evolving for long enough in a steady, longitudinally uniform, turbulent flow field Taylor (1954) showed that the ADM applies to the cross-sectional average concentration of a conservative solute and that the magnitude of the dispersion coefficient is controlled by the details of the velocity and mixing fields. His model (originally derived for flows in pipes) is described by the following equation:

$$\frac{\partial c(x,t)}{\partial t} + U_{AD} \frac{\partial c(x,t)}{\partial x} = D_{AD} \frac{\partial^2 c(x,t)}{\partial x^2}$$
 (1)

where c is the cross-sectional average solute concentration,  $U_{AD}$  is the cross-sectional average longitudinal flow velocity,  $D_{AD}$  is the dispersion coefficient, t is time and x is the longitudinal co-ordinate direction.

Fischer (1967) showed that since rivers have large aspects ratios the transverse variations in longitudinal velocity and cross-sectional mixing are much more important than the corresponding vertical variations. Consequently much use is made of vertically averaged flow and mixing parameters when dispersion coefficients in rivers are estimated from flow and mixing parameters, see e.g. Rutherford (1994); Wallis and Manson (2004). An advantage of the ADM is access to easily applied analytical solutions of Eq. (1). On the other hand these solutions do not always fit observations well.

# 2.2 The Transient Storage Model (TSM)

The TSM is an extended version of the ADM, and its origins can be traced back over about 50 years. The earliest need to modify the ADM stemmed from work in pipes and channels which suggested that the storage and slow release of solute by laminar boundary layers might explain some discrepancies between observations and model predictions (Taylor 1954; Elder 1959). In rivers a similar mechanism was attributed to dead zones which were originally associated with "pools and stagnant areas, or both, cause by debris or unevenness of the banks or bottom" (Thackston and Krenkel 1967). Further evidence from rivers, see e.g. Nordin and Sabol (1974) and Day (1975), suggested that observed concentration-time profiles were not predicted well by the ADM. More recently, several other geomorphologic features, such as pool-riffle structures (e.g. Bencala and Walters 1983) and interactions between a river channel and the surrounding hyporheic zone (e.g. Elliott and Brooks 1997) have been added to dead zones, and the term transient storage is now often used to describe the effects of a range of storage phenomena on solute transport in rivers (Worman 2000; Wallis et al. 2013).

The TSM is described by two equations, one representing solute transport in the main river channel, including advection, dispersion and the effect of the storage zones, and the other representing a dynamic mass balance of solute in the storage zones themselves. In these equations a first-order exchange mechanism is used to describe the transport of solute between the main channel and the storage zones (and back again) and the solute is assumed to behave conservatively. Several slightly different formulations of the equations have appeared in the literature since their first appearance (Thackston and Krenkel 1967), see e.g. Bencala and Walters (1983), Rutherford (1994), Worman (2000), Deng et al. (2010), Bottacin-Busolin et al. (2011) and Wallis et al. (2013). These reflect various issues such as the choice of geometric variables used, the inclusion of lateral inflow, the interpretation of the transient storage process(es) and the presence of variable time scales over which the transient storage takes place. The model equations used in this work, assuming steady and longitudinally uniform flow, are:

$$\frac{\partial c(x,t)}{\partial t} + U_{TS} \frac{\partial c(x,t)}{\partial x} = D_{TS} \frac{\partial^2 c(x,t)}{\partial x^2} + k_1 (s(x,t) - c(x,t))$$
 (2)

$$\frac{\partial s(x,t)}{\partial t} = -k_2(s(x,t) - c(x,t)) \tag{3}$$

where  $U_{TS}$  is the cross-sectional average flow velocity in the main channel,  $D_{TS}$  is the dispersion coefficient in the main channel,  $k_1$  and  $k_2$  are model parameters (see below), s is the solute concentration in the storage zones and the other symbols are as previously defined. Note that the velocity and dispersion appearing in Eq. (3) are not necessarily the same as the corresponding parameters appearing in Eq. (1).

The model parameters introduced above are defined as:

$$k_1 = \alpha \tag{4}$$

$$k_2 = k_1 \frac{A}{A_S} \tag{5}$$

where  $\alpha$  is the exchange rate between the main channel and the storage zones, A is the cross-sectional area of the main channel and  $A_S$  is the cross-sectional area of the storage zones. The model is often successful in fitting observations, but the interpretation of the transient storage parameters is not straightforward, particularly when different combinations of parameter values yield very similar outputs.

# 2.3 The Aggregated Dead Zone Model (ADZM)

Similarly to the origins of the TSM, the ADZM was developed because of reported deficiencies of the ADM. In a radical departure from previous work, it was postulated (Beer and Young 1983) that the dispersion occurring in dead zones

dominated the shear flow dispersion. Hence a model could be constructed on the basis of transient storage only. Or more pragmatically, the effects of all dispersive mechanisms in a river reach could be amalgamated and represented by a single effective dead zone. A further radical approach was to formulate the model only in the time domain (by spatial integration of the physical processes). This had several consequences, e.g. a much simpler mass balance equation than either the ADM or TSM and access to powerful model calibration techniques via time-series analysis (Young 1984).

The model resembles simple hydrologic models for flood propagation in rivers, many of which are based on the concept of storage routing (Young and Wallis 1985; Shaw et al. 2011). Importantly, however, the ADZM includes an explicit time delay to cater for the purely advective transport processes. The transport of a conservative solute in steady flow is described by the following equation:

$$\frac{\partial y(t)}{\partial t} = \frac{1}{V} \left[ Q_u u(t - \tau) - Q_y y(t) \right] \tag{6}$$

where y(t) and  $u(t-\tau)$  are the cross-sectional average solute concentrations at the downstream and upstream ends of a river reach, respectively,  $\tau$  is the time delay, V is the volume of the aggregated dead zone and  $Q_y$  and  $Q_u$  are, respectively, the flow rates at the downstream and upstream ends of the reach. In practical terms:  $\tau$  is the minimum reach travel time (time interval between the first arrival of solute at the two ends of the reach);  $T_{ADZ}$  ( $V/Q_y$ ) is the time solute spends in the aggregated dead zone; and the sum of  $\tau$  and  $T_{ADZ}$  is the ADZ travel time. Wallis (1994) showed that ADZ travel time is theoretically equal to the time interval between the centroids of temporal solute concentration profiles at the two ends of a reach.

Several studies have shown that the ADZM is able to reproduce solute transport in rivers very satisfactorily (Wallis et al. 1989; Green et al. 1994; Lees et al. 2000). However, the fact that the model's parameters are not related to the traditional concepts of advection and dispersion may be considered a disadvantage (Rutherford 1994). On the other hand, both the time delay and the residence time for a reach have been found to vary with flow rate in a physically realistic way, both decreasing as flow rate increases (Wallis et al. 1989; Green et al. 1994).

#### 3 Data Collection

For each experiment, YSI-600xlm multiparameter sondes (YSI, Yellow Spring, USA) were placed at two longitudinal stations (typically about 60 m apart) in the study stream and set to record conductivity at a fixed time interval (typically around 2–10 s). Pre-weighed NaCl was fully dissolved in a small amount (typically 2 L) of stream water and then immediately released into the stream at some distance upstream of the upper station. Generally the initial mixing zone was sufficiently long (typically about 10–20 m) for complete cross-sectional mixing to take place

before the upper station. In the shortest reaches, additional deflectors and pools were created upstream of the upper station to increase mixing. Any naturally occurring background conductivity signal was subtracted from the observations prior to the modelling. The flow rates at the stations were evaluated via dilution gauging, and the upstream and downstream temporal conductivity profiles were analysed to elucidate stream transport parameters, as described below. The median (range) stream characteristics were: width 0.9 (0.2–3.6) m, depth 6 (1–23) cm, velocity 15 (1–52) cm s<sup>-1</sup>, reach length 36 (13–107) m and flow rate 7 (0.2–70) L s<sup>-1</sup>.

## 4 Application of Models

Equation (1) and equation system (2/3) were solved using a finite volume approach in space, evaluating the advection term explicitly in time and evaluating the dispersion and transient storage terms implicitly in time. The DISCUS method (Wallis et al. 1998; Manson and Wallis 1999; Manson and Wallis 2000; Manson et al. 2001) was used for the advective terms in Eqs. (1) and (2) and the Crank-Nicolson method (Press et al. 1992) was used for the dispersion terms in Eqs. (1) and (2) and for the transient storage term in Eq. (2). The rationale behind these choices is that both methods are individually well-suited, being unconditionally stable and robust, for those particular terms, respectively. Equations (3) and (6) were solved using the Crank-Nicolson method.

The models were fitted to the observations by parameter optimization, minimising the sum of squared residuals, SSR, by a modified Levenberg-Marquardt algorithm (Press et al. 1992). A Python script was used to undertake the minimisation by calling C code implementations of the three models. Python and C were linked using the BOOST library. Only main channel concentration was used for the fitting because concentrations in the storage zones were not measured. A normalised fitting parameter (NRMSE) was defined for comparing the performance of the models to different data sets, defined by:

$$NRMSE = \frac{1}{\max(C_{OBS})} \left(\frac{SSR}{N}\right)^{0.5} \tag{7}$$

where N is the number of data points and  $C_{OBS}$  refers to the observed concentration profile. To aid the identification of the best model for any individual data set, allowing for a penalty for using more parameters than were justified, the Akaike Information Criterion (AIC) was calculated (assuming the Gaussian case) using:

$$AIC = 2k + N \ln\left(\frac{SSR}{N}\right) \tag{8}$$

where k is the number of estimated parameters, including the residual error term (Burnham and Anderson 2002). The best model has the smallest AIC. For the TSM  $\alpha$  and  $A_s/A$  were evaluated from the optimized values of  $k_1$  and  $k_2$  using Eqs. (4) and (5). The effect of lateral inflow was included in the ADM and TSM by adding the term -qc(x,t)/A to the right-hand sides of Eqs. (1) and (2). The lateral inflow rate, q, was known from the upstream and downstream flow rates and A was estimated from the flow rates and the centroid velocity (see Sect. 5).

#### 5 Results and Discussion

All three models were successfully optimized to the great majority of the 70 sets of tracer data. A few cases failed (5 with the ADM, 9 with the TSM, 5 with the ADZM) either due to inherent problems with the data or due to convergence problems during optimization. Results are presented in various ways below. Figure 1 shows the goodness of fit of the successfully optimized models to all the data. The mean goodness of fit for each model across all the data is also shown.

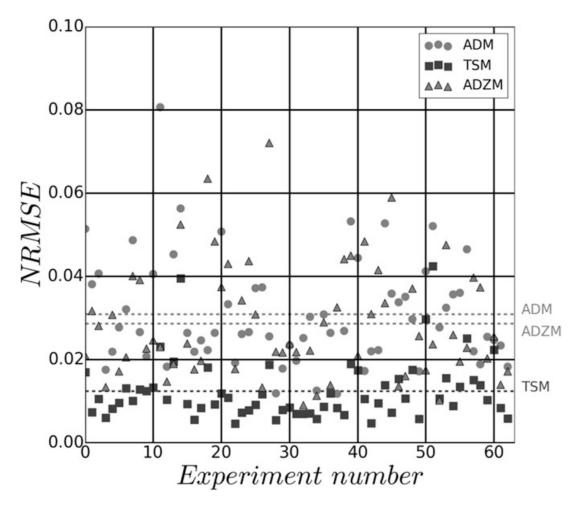


Fig. 1 Comparison of goodness of fit

Although there is a lot of scatter, it is clear that the TSM tends to fit the data best (mean NRMSE of  $\sim 0.01$ ). The ADE and ADZ models achieve a similar average goodness of fit to each other (mean NRMSEs of  $\sim 0.03$ ), which is significantly larger than the mean value for the TSM. This is expected because, with 4 parameters, the TSM has a greater ability to represent the finer detail of the solute transport processes than either of the two 2-parameter models. However, it is important to guard against the possibility of over-fitting. Hence values of the AIC were also considered. These showed that in nearly all cases the order of model preference was TSM, ADZM and ADM. In a few cases, corresponding with little transient storage activity, the ADM was preferred to the other two models.

Figure 2 shows correlations between logarithms of the parameters of the TSM and the ADM. Arbitrarily assuming that a correlation coefficient (absolute

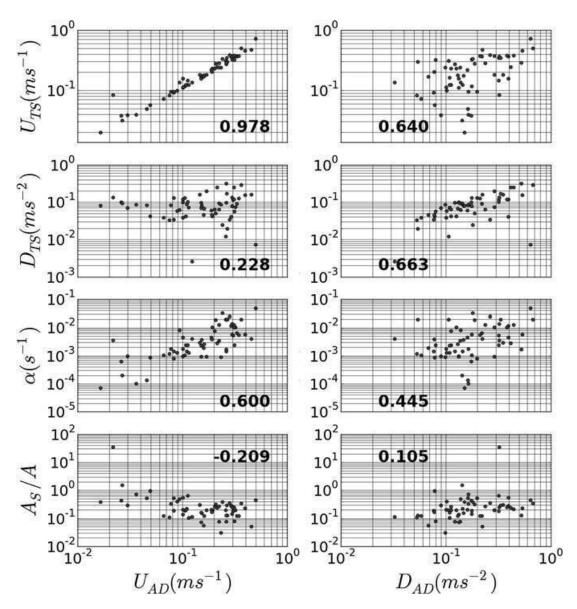


Fig. 2 Comparison of TSM and ADM parameters (with correlation coefficients)

value) >  $\sim 0.6$  indicates a strong correlation, relationships exist between  $U_{TS}$  and  $U_{AD}$ ,  $\alpha$  and  $U_{AD}$ ,  $U_{TS}$  and  $D_{AD}$ , and  $D_{TS}$  and  $D_{AD}$ . The correlations between the two velocities and the two dispersion coefficients are expected. In general  $U_{TS} > U_{AD}$  because in the ADM the retardation of longitudinal tracer transport due to transient storage can only be accounted for by  $U_{AD}$ . Similarly, in general  $D_{TS} < D_{AD}$  because in the ADM dispersion caused by transient storage can only be accounted for by  $D_{AD}$ . The range of velocity values  $(0.02-0.5 \text{ ms}^{-1})$  is a consequence of the different slope, roughness and flow rate of the streams, and the range of the dispersion coefficients  $(0.03-0.7 \text{ m}^2\text{s}^{-1})$  is typical for the stream size (Heron 2015).

The correlation between  $\alpha$  and  $U_{AD}$  is consistent with the idea that the boundary layer across which solute exchange takes place decreases as velocity increases.

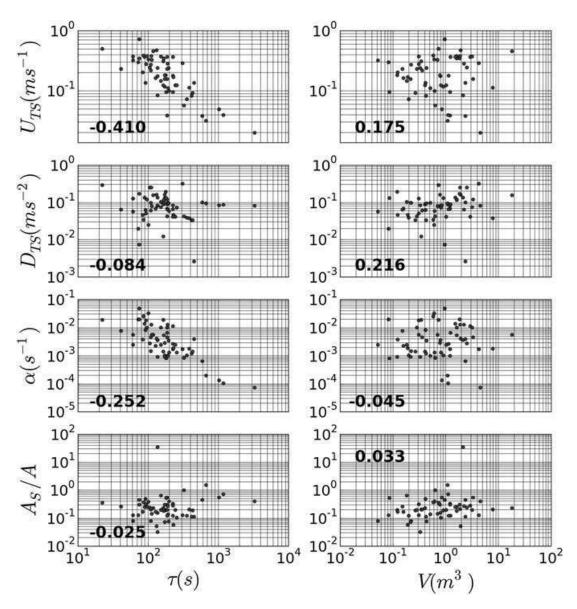
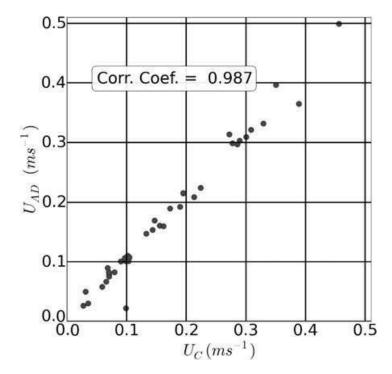


Fig. 3 Comparison of TSM and ADZM parameters (with correlation coefficients)

**Fig. 4** ADM velocity vs centroid velocity

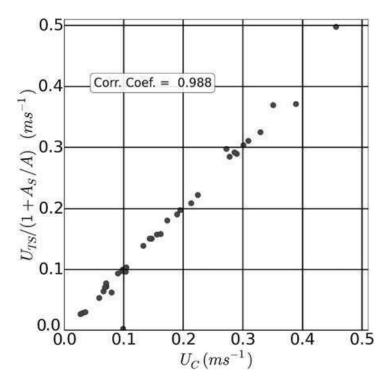


Although it is interesting that there appears to be a relationship between  $U_{TS}$  and  $D_{AD}$ , it is of little practical value since  $U_{TS}$  is more strongly correlated with  $U_{AD}$ . Values of  $\alpha$  and  $A_s/A$  are typical of those found in previous applications of the TSM to mountain streams, see e.g. D'Angelo et al. (1993) and Gooseff et al. (2003).

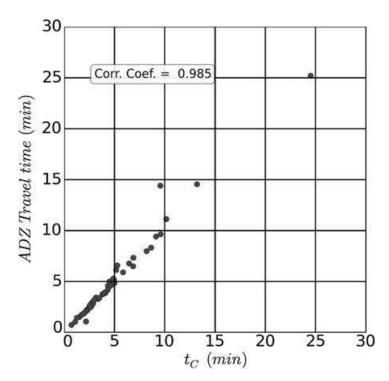
Using the same criterion as above for identifying strong correlations, Fig. 3 suggests there are no relationships between the logarithms of the parameters of the TSM and the ADZM. Interestingly, however, two of the largest correlation coefficients are between  $U_{TS}$  and  $\tau$  and  $\alpha$  and  $\tau$ . Since we might expect time delay to be correlated with velocity, these are consistent with the relationships identified from Fig. 2. Finally, it is worth reporting that the range of values of the ratio of residence time to reach mean travel time ( $T_{ADZ}/(\tau + T_{ADZ})$ ), known as the dispersive fraction, (0.1–0.6) is consistent with previous work (Wallis et al. 1989; Green et al. 1994; Guymer 2002).

Figures 4, 5 and 6 compare various advective transport parameters against the movement of the centre of mass of the solute cloud. The latter was evaluated from the centroids of the upstream and downstream temporal tracer concentration profiles, giving estimates of centroid travel time,  $t_C$ , and centroid velocity,  $U_C$ . Both of these are independent of the ADM, TSM and ADZM. Figure 4 suggests a one-to-one relationship between  $U_{AD}$  and  $U_C$ , indicating that the ADM correctly describes the bulk transport of the solute cloud. Figure 5 shows a modified TSM velocity plotted against  $U_C$ . The modification reduces  $U_{TS}$  by an amount that reflects the effect of the transient storage (Czernuszenko and Rowinski 1997; Worman 1998; Lees et al. 2000). The figure suggests that the combination of the optimized TSM parameters correctly describes the bulk transport of the solute cloud. Figure 6 shows the ADZ travel time plotted against  $t_C$ . Again the

Fig. 5 Modified TSM velocity vs centroid velocity



**Fig. 6** ADZ travel time vs centroid travel time



combination of the optimized parameters correctly describes the bulk transport of the solute cloud. Overall, the good agreement between these velocities and travel times suggests that the optimized parameter values are robust.

#### 6 Conclusions

Three solute transport models were optimized to a large number of sets of tracer data collected in similar mountain streams. The TSM consistently fitted the tracer data better than either the ADM or the ADZM: the ADZM performed a little better than the ADM. Apparently useful correlations were found between the logarithms of some of the parameters of the TSM and the ADM, but no useful correlations were found between the parameters of the TSM and the ADZM. For all three models, combinations of the optimized parameters correctly described the bulk movement of the solute cloud, giving confidence in the optimized parameters.

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