Temporal variability in mean transit time and transit time distributions assessed by a tracer-aided tank model of a meso-scale catchment

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Abstract:

The steady-state assumption for catchment transit time is a controversial issue in catchment hydrology. In this study, we propose a new approach to estimate the time-variant mean transit time (MTT) and transit time distribution (TTD) using a five-layer tank model with isotopic tracers and test it in the Fuefuki River catchment, central Japan. Model parameters were optimized during the calibration phase based on hydrometric and isotopic observations and then validated in a separate validation phase. The long-term (2003–2012) average MTT was estimated to be 23.7 years. However, the daily MTT was highly variable, ranging from 1.2 to 37.0 years. Instantaneous TTD also varied markedly. Precipitation alters TTD by increasing younger components and shortens the MTT. Thus, a steady-state assumption is inappropriate, with the relationship between monthly MTT and precipitation amount most closely approximated by an exponential function. The dependence of MTT on precipitation is an important descriptor for characterizing catchments. Although optimized model-parameters have some uncertainties, potential errors in estimating the MTT are relatively small (e.g., < ±3 years). Therefore, the tracer-aided tank model is useful for estimating time variations in MTT and TTD with high reliability.

KEYWORDS transit time; tank model; isotope tracer; catchment hydrology; Fuefuki river

INTRODUCTION

Transit (or travel) time is defined as the elapsed time from when a water molecule enters a system (or reservoir; e.g., catchment, aquifer, lake) until it exits (Bolin and Rodhe, 1973). The catchment transit time is a lumped descriptor that reflects storage, flow pathways and sources of water in a catchment (McGuire and McDonnell, 2006). It allows us to examine how catchments retain and release water that is contaminated, for example, by chemical/nuclear accidents. Here, it should be noted that transit time differs for each individual molecule. Thus, we can define the mean transit time (MTT) and transit time distribution (TTD) for a mass of water collected at a given time and location (i.e., outlet of the catchment), where MTT is the mean of transit times for all water molecules (rather than a temporal average) and TTD the distribution of transit time of each molecule within the mass of water.

In earlier work on transit time using environmental tracers (Maloszewski and Zuber, 1982; Maloszewski et al., 1983; DeWalle et al., 1997; Ozyurt and Bayart, 2003), a number of idealized, hypothetical TTD functions using a steady-state assumption were employed to estimate MTT. The same or similar, time-invariant TTD functions were applied in unsteady-state (i.e., variable flow) cases for reproducing tracer concentration variations (Zuber, 1986) and for estimating temporally averaged MTT (Ozyurt and Bayart, 2005). Amin and Campana (1996) proposed hypothetical, time-variant TTD functions but these were not tested with observed data. In a field experiment where a step change in tracer concentration was applied across a small catchment, Rodhe et al. (1996) estimated a non-idealized TTD. Lindström and Rodhe (1986) used a lumped hydrological model to simulate temporal variation of observed tracer concentration in stream water and the TTD was then obtained by applying a virtual tracer pulse to the model with steady-state boundary conditions. Recently, McGuire et al. (2002) predicted time-variant TTD under field conditions using a simple hillslope model calibrated with hydrometric and tracer observations. Botter et al. (2010) also obtained a time-variant TTD using a hillslope-scale stochastic soil moisture model with variable boundary conditions. Furthermore, Sayama and McDonnell (2009) developed a time-space accounting scheme with a distributed hydrologic model for tracking not only temporal but also spatial sources of runoff components. These recent contributions to the literature demonstrated that MTTs were not always constant and that temporally averaged TTDs had a near-exponential form with a short time-lag. However, the models used in these studies were highly complex, with many uncertain model-parameters, so the reliability of the MTT and TTD estimates remains unclear. In addition, many of these studies were limited in that they were only applied to small catchments.

In the present study, we propose and test an approach to estimate time-variant MTTs and TTDs with a relatively simple, lumped hydrological model that was calibrated and validated by hydrometric and isotopic tracer observations to reduce uncertainties in transit time estimations. Our objectives were twofold: (1) to clarify the temporal variation characteristics of MTTs and TTDs in a meso-scale catchment for re-examining the steady-state assumption, and (2) to evaluate uncertainties in the estimated MTT. The reason we focused on meso-scale (approximately 105–107 km2; Uhlenbrook et al., 2004) catchments is because these are more strongly linked to the humanosphere than small, remote
catchments. The outcomes of the study provide insights into
catchment hydrology and biogeochemical cycles and will be
useful for managing chemical/nuclear disaster risks.

MATERIALS AND METHODS

Study site and data set

The study catchment was the Fuefuki River catchment
(35.5°–35.9°N, 138.5°–138.9°E), in the northeastern part of
the Fuji River basin, central Japan (Figure 1). The area of
the catchment is 905.70 km² and elevation ranges from
approximately 250 to 2600 m. Northern, eastern and
southern parts of the catchment are characterized by
mountainous topography, while central and eastern parts are
alluvial fans and lowlands. Mountains are formed mostly
by granite and partly by andesitic/basaltic rocks. The alluvial
fans are covered by gravel with a thickness of 20–30 m,
underlain by a clay layer of similar thickness. According to
the meteorological observation records at the Kofu Station
of the Japan Meteorological Agency (JMA), the climatic
norm (1981–2010) for annual precipitation is 1135.2 mm,
with a mean temperature of 14.7°C, mean relative humidity
of 65%, and mean wind speed of 2.2 m/s. The dominant
land use/land cover type is forest in the mountainous areas,
orchard and vegetable fields in the alluvial fans, and
residential areas and paddy fields in the alluvial lowlands.

The study catchment was defined considering the
location of a gauging station (Y3 in Figure 1) maintained
by the Ministry of Land, Infrastructure, Transport and
Tourism (MLIT), Japan. Discharge rates in the river are
routinely observed at this station by MLIT and observed
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Tourism (MLIT), Japan. Discharge rates in the river are
routinely observed at this station by MLIT and observed
data are open to the public. We performed the first monthly
monitoring at three locations (KOF, ANY and SAN in Figure
1; May 2010–June 2012) and semi-weekly (i.e., twice per
week) monitoring at one location (KOM in Figure 1; May
2011–Apr. 2012) were also carried out. Hydrogen and
oxygen stable isotope ratios ($^{2}H$ and $^{18}O$) of collected
water samples were measured with a liquid water isotope
analyzer (L1102-i, Picarro, CA, USA). Measured results are
expressed using δ notation (i.e., δ$^{2}H$ and δD) relative to
the Vienna Standard Mean Ocean Water (V-SMOW).
Measurement errors for the analyzer were 0.1‰ for δ$^{18}O$
and 1‰ for δD (Yamanaka and Onda, 2011).

The Radar-AMEDAS (Automated Meteorological Data
Acquisition System) precipitation data (e.g., Makihara et al.,
1996) produced by the JMA were used as model input for
the period of 2006–2012. Data before 2005 were not used,
because the spatial resolution is not the same as after 2006.
For the purposes of calibration and validation of the model,
we used daily river discharge data for the period 2006–2010,
since data after 2011 was not available. Isotope monitoring
data mentioned above were also employed for calibration/
validation of the model. In addition, meteorological data
including point precipitation data observed at Kofu and
adjacent stations of JMA were used for evapotranspiration
estimation and long-term (2003–2012) estimation of MTT.

Model

The so-called “tank model” (Sugawara, 1995), a widely
applied rainfall-runoff model (Brutsaert, 2005), was
employed for estimating time-variant MTT and TTD. While
the tank model is a lumped (i.e., non-distributed) conceptual
model, it can consider as having different flow paths and
water pools with different, variable transit times. It has been
used not only for predicting runoff but also for simulating
temporal changes in water quality (Kato, 2005). Ikawa et al.
(2011) also successfully reproduced temporal variations of
observed isotopic compositions for stem flow using a two-
layer tank (i.e., canopy tank and stem tank) model. In the
present study, a five-layer tank model was developed to
conceptually represent overland flow, rapid throughflow,
delayed throughflow, groundwater flow and bedrock flow
(Supplement Figure S1). In addition to water flow, isotope
transport was also computed taking into account the isotopic
variability of precipitation input, isotopic fractionation
during soil evaporation and isotopic mixing within each
tank. A daily time step was adopted for both water flow and
isotope transport computations.

Detailed description (e.g., basic equations and treatment
of evapotranspiration processes) of the model is given in
Supplement Document S1. Although the model itself is not
new, this study is the first application of a tank model that
is calibrated/validated not only by hydrometric observation
but also by isotopic tracers to obtain reliable estimation of
time-variant MTT and TTD. Values of model parameters
were determined during the calibration phase (2006–2007
for water flow, and May 2010–July 2011 for isotope
transport) by a trial and error method, considering the Nash-
Sutcliffe efficiency (NSE; Nash and Sutcliffe, 1970) for
water flow and root mean square error (RMSE) for isotope
transport. Validation of the model was performed in separate,
validation phases (2008, 2009 and 2010 for water flow, and
Aug. 2011–Mar. 2012 for isotope transport). The periods of
calibration and validation were determined considering the
length of available data for each of the water flow and
isotope transport simulations. Ideally, the calibration period
requires more than one year due to seasonal and inter-annual variations. Unfortunately, the isotope monitoring period was less than two years, so the first 60\% of the period was set as the calibration phase and the rest as the validation phase.

**Estimation scheme for time-variant MTT**

To estimate time-variant MTT using a calibrated/validated tank model, we introduce a virtual (or imaginary) “age” tracer into the model. The virtual-age-tracer approach has been already attempted by Goode (1996) for groundwater and Khatriwala et al. (2013) for ocean water.

The concentration of this conserved, nonreactive age tracer, $A(i)$, is computed by

\[
\frac{dh(i)A(i)}{dt} = q_H(i-1)A(i-1)
\]

\[-[f_T(i)T_r + q_T(i) + q_H(i)]A(i) - f_E(i)E_S A(i) + 1(1)\]

where $h(i)$ is the water level in the $i$-th tank, $q_H(i)$ is the vertical water flux, $q_T(i)$ is the horizontal (exactly speaking, toward a stream network) water flux, $T_r$ is the transpiration, $E_S$ is the soil evaporation, and $f_T(i)$ and $f_E(i)$ are the weighting factors at the $i$-th tank for root water uptake and soil evaporation, respectively. If we define the age as the elapsed time after the water enters the catchment across the ground surface, then $A(1) = 0$ throughout the simulation period. Solving Equation (1) for $A(i)$ under this boundary condition gives the mean age of water in each tank. Flux-weighted mean of age for total runoff ($A_Q$), which is equivalent to MTT, can then be obtained as

\[
A_Q = \frac{\sum_{i=1}^{5} q_H(i)A(i)}{Q}
\]

(2)

where $Q$ is the total runoff. If we take a time step of 1 day, the unit for $A(i)$ and $A_Q$ is days; and the last term of Equation (1), that is unity, indicates the rate of ageing.

A common issue for previous non-steady-state approaches is assigning the initial conditions. In the present study, initial values of $A(i)$ were first assumed to be zero for all tanks, and then temporal evolution of $A(i)$ was computed for several years as a spin-up run.

**Estimation scheme for time-variant TTD**

The scheme for predicting time-variant TTD is independent of that for MTT. In this scheme, a virtual (or imaginary) “date” tracer is introduced to the model as a pulse (or artificial) input at a time window rather than as continuous (or natural) input. We next consider the concentration of the date tracer, $D(i, j)$, where $i$ is the tank number and $j$ is the number of the time window for tracer input. Although the length of the time window is arbitrary, we adopted 30 days in the present study. The $D(1, j)$ is assigned to be unity for the time window: $30(j-1) < t < 30j$. Temporal evolution of $D(i, j)$ at each tank was then computed using the following equation:

\[
\frac{dh(i)D(i)}{dt} = q_H(i-1)D(i-1)
\]

\[-[f_T(i)T_r + q_T(i) + q_H(i)]D(i) - f_E(i)E_S D(i)\]

(3)

Also, we can compute the concentration of the date tracer in total runoff water, $D_Q(j)$, as:

\[
D_Q(j) = \frac{\sum_{i=1}^{5} q_H(i)D(i, j)}{Q}
\]

(4)

The value of $D_Q(j)$ at a given time represents the proportion of water that entered below ground at the $j$-th time window compared with the total runoff water at that time. Finally, TTD can be obtained by slicing the time series of $D_Q(j)$ for different $j$ at an arbitrary time and rearranging it along $j$.

**RESULTS AND DISCUSSION**

Model performance for water flow and isotope transport

Water flow simulation generally showed good reproducibility with partial underestimation (e.g., 2006 and a former period of 2010) and overestimation (e.g., from mid-2007 to the end of 2008) of runoff (Figure 2a). Although NSE was higher during the calibration phase (0.308) than in the validation phase (0.149 in 2008, 0.184 in 2009, and 0.223 in 2010), year-to-year variation was large. Such variability in accuracy may be attributed to not only incompleteness of the model but also the quality of the observation data for both river discharge and precipitation.

Isotopic variation in river water was also generally well reproduced with some minor deviations (Figure 2b). In the calibration phase, RMSE was 0.23\% for $\delta^{18}O$ and 2.1\% for $\delta D$. While the values in the validation phases were slightly worse (0.35\% for $\delta^{18}O$ and 2.7\% for $\delta D$), these values are sufficiently low compared with the measurement accuracy of the isotope analyzer. In isotopic transport simulation, monthly data for the precipitation isotopes were applied until April 2011, after which semi-weekly data were applied in

![Figure 2. Comparison of (a) hydrograph and (b) river water isotopic composition, between observation and simulation throughout calibration and validation phases](image-url)
order to assess the effect of the temporal resolution of precipitation isotope data on simulation results. No significant difference could be found in simulated river water isotope composition due to changes in the temporal resolution of precipitation isotope data. This suggests thorough mixing of old and new precipitation within shallow soil layers over the time scale of a week to a month.

Mean transit time

The long-term (Oct. 2003–Sep. 2012) average of MTT was estimated to be 23.7 years. However, estimated daily MTT showed high variability, ranging from 1.2 to 37.0 years (Figure 3). This variation is apparently associated with variation in the precipitation amount, with a correlation coefficient ($R$) of 0.45, indicating that river water at the catchment outlet becomes markedly younger through precipitation events. In addition, it is worth noting that MTT is not stable over time, even during rainless periods, suggesting that the MTT of so-called “base flow” cannot be regarded as constant.

Taking monthly average values of MTT, the relationship between MTT and precipitation amount can be closely approximated by an exponential function with a determination coefficient ($R^2$) of 0.73. The change in MTT per unit increase in monthly precipitation was lower in months with greater precipitation (Figure 4). This fact suggests that greater precipitation events enhance not only shallow water flow but also deeper water flow, with the MTT being temporally more stable in the latter case.

Clearly, the model developed in the present study is far from perfect and oversimplifies hydrological processes in the real world to a greater or lesser extent. Therefore, we must assess the reliability of the estimated MTT. Figure 5 shows the results of 100 Monte Carlo simulations using different randomly selected model-parameters within the range of $\pm 5\%$ of optimal values (McGuire et al., 2007). The deviation of estimated MTT from the optimal solution ($\Delta$MTT) ranged within $\pm 3$ years, demonstrating that potential errors in MTT estimation due to uncertainties in the model parameters were relatively small. In addition, the error tended to decrease in cases of higher $NSE$ and lower combined-$RMSE$ (where combined-$RMSE$ is defined as a sum of $RMSE$ for $\delta^{18}O$ and one eighth of $RMSE$ for $\delta D$). This suggests that optimal values of the model parameters can be calibrated well using $NSE$ and $RMSE$, with high reliability.

Many prior studies from around the world have reported MTTs of several years or shorter (McGuire and McDonnell, 2006). However, Michel (1992) reported MTTs longer than 10 years for very large catchments (> 10,000 km$^2$). In addition, using chlorofluorocarbons and Si concentration, Ohara (2009) estimated MTT for areas adjacent to the present study catchment to be 6–19 years. These values are comparable with our results. Thus, we are confident in the validity of the estimated MTT, although it is still unknown whether the relatively longer MTT can be attributed to the spatial scale or the topographic/hydro-geological features of the study catchment.

Transit time distribution

Two examples of estimated TTD are shown in Figure 6. The first case (4 Sep 2011) reflects situations just after a large storm event. The peak in transit time is located on the left-hand edge; the youngest component (i.e., transit time < 30 days) is the most dominant contributor, accounting for more than 20% of total river water. The shape of the TTD is similar to an exponential form, while we can find 1-yr periodic variations, probably due to seasonal variations in precipitation. Conversely, the second case (17 Feb 2011) reflects low flow conditions during a long drought period. The peak in transit time shifts toward a greater (i.e., older) time domain and the peak height is small (< 0.5%). The
shape of the TTD for this case is similar to neither exponential nor any other known functions. Consequently, it is clear that the TTD is highly variable over time, reflecting that the history of precipitation variation, and the assumption of idealized TTD functions, which have often been applied in previous studies, are not appropriate.

In the present study, TTD computation was performed for six years (2006–2011), so that components older than six years (= 2,190 days) cannot be explicitly evaluated. Because the MTT extends to more than 20 years, Figure 6 does not show the entire range in TTD. In other words, integration of the TTD shown in Figure 6 cannot give the actual MTT. Therefore, the use of a TTD estimation scheme is not recommended for the purpose of MTT estimation.

To obtain time-variant TTDs, McGuire et al. (2007) employed a similar approach. However, they evaluated it as temporal variation of a tracer concentration that was injected instantaneously at a given time in the model; it corresponds to a single $D_Q(j)$ time-series in our scheme. In this context, the approach of Botter et al. (2010) is the same. In contrast, we evaluated TTD by combining multiple $D_Q(j)$ time-series, similar to Sayama and McDonnel (2009). Although this procedure is somewhat complicated, it should represent the TTD more accurately for water sampled at a given time.

CONCLUSIONS

For the Fuefuki River catchment, the long-term average of MTT was estimated to be 23.7 years. This is relatively long compared with values reported in previous studies for mountainous, small catchments. Nevertheless, the daily MTT was highly variable. In addition, the TTD was also variable and an exponential model was applicable only during or just following large storm events. Precipitation is a dominant causative factor for temporal variability in MTT and TTD. Precipitation alters TTD, with an increase in younger components, and shortens MTT. In other words, river water is repeatedly rejuvenated by precipitation and stemflow under a Japanese cedar forest during a typhoon event. Hydrological Research Letters 5: 32–36. doi: 10.3178/hrl.5.32.

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