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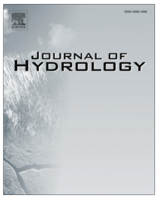

Editorial

Residence times in subsurface hydrological systems, introduction to the Special Issue

Interest in the residence time distribution (RTD) as a comprehensive measure of subsurface hydrologic systems is growing. This focus is resulting from recognition that diverse vadose zone, groundwater flows, and transfer between hydrological compartments, are fundamentally related to the system RTD. Furthermore, transport of chemical or biological species and the biogeochemical activities that govern their fate, is principally reflected by the system RTD. Thus the RTD is used in geochemical interpretation of environmental tracers, in direct reactive transport approaches, and ultimately for sustainability and protection assessments in the consideration of transient boundary flows due to climate change or other causes, anthropogenic and/or natural. The RTD has been handled in the past primarily as a byproduct of models. It is now increasingly viewed as an integrative characteristic for which shape-free and generic distributions are developed, that links conceptual hydrology, characterization data, and mathematical models. Intermediary between mechanistic modeling, geochemical data and predictions, the role for residence time distribution is to represent consistently the flow, transport and reactivity processes while reaching the objective of biogeochemical interpretation and sustainability assessment. After some outline of the scientific context, we introduce the contributions of this special issue and conclude with the emerging challenges.

1. Context

Residence times in subsurface systems have been shown to display wide variabilities: of weeks to years close to the surface for interflow, throughflow or subsurface storm flow (McDonnell et al., 2010; McGuire and McDonnell, 2006; Rinaldo et al., 2011; Tetzlaff et al., 2009); of months to centuries for unconfined aquifers (Cook and Herczeg, 2000; Kazemi et al., 2006; Leibundgut et al., 2009); reaching 10^3–10^5 years for deeper confined systems (Bentley et al., 1986; Bouchez et al., 2015; Glynn and Plummer, 2005; IAEA, 2013; Plummer et al., 2004; Sturchio et al., 2004). To a lesser extent, residence times vary temporally following hydrological fluctuations (Duffy, 2010; Freyberg, 1986; Harman, 2015; Harvey et al., 2006), anthropogenic modifications of boundary conditions and source/sink terms (Bexfield and Jurgens, 2014; Bowman and Rice, 1986; Broers and van der Grift, 2004; Zinn and Konikow, 2007b), and climatic changes (Goderniaux et al., 2013; Klove et al., 2014). Residence time variability is not only observed spatially and temporally but also within single samples as evidenced by the simultaneous presence of multiple tracers of highly different temporal signatures (Genereux et al., 2009; Heilweil et al., 2009; Jasechko, 2016; Kashiwaya et al., 2014; Koh et al., 2006; Labasque et al., 2014; Solomon et al., 2010). Even though measurement errors are difficult to systematically discard, the simultaneous occurrence of widely different tracers is rather thought to arise from multiple mixing mechanisms (Castro and Goblet, 2005; Land and Timmons, 2016), some of which may be induced by the sampling act itself, e.g., borehole fluid mixing. Progress in the understanding of fundamental mixing processes has continuously influenced the conceptualization of residence times and the associated interpretation of tracer data.

While piston-flow models and associated Dirac RTDs have been extensively used as a direct way to straightforwardly interpret concentrations of atmospheric tracers in terms of temporal measures generally called "groundwater ages" (Begemann and Libby, 1957; Suckow, 2014), broad RTDs seem to be the rule rather than the exception. Distribution of water ages within samples arises fundamentally from the multi-scale nature of spreading and mixing in geological media (Dentz et al., 2011; Gelhar et al., 1992). Both prevail from the pore scale to the formation scale. From the pore scale to the Darcy scale, effects of local velocity fluctuations are classically modeled by equivalent hydrodynamic dispersion concepts (Bear, 1973; Zheng and Bennett, 2002), which quickly transform the Dirac distribution in wider inverse Gaussian-like transit-time distributions (Ogata and Banks, 1961; Wexler, 1992). Times are even more broadly distributed when accounting for chemical sorption and physical trapping mechanisms in low flow zones dominated by diffusive transport (Carrera et al., 1998; Haggerty and Gorelick, 1995) with possible power-law distributions of the late transit times (Berkowitz and Scher, 1997; Dentz et al., 2004). Such kind of non-Fickian transport has been well identified and transferred to the groundwater dating community and interpretation models have been adapted accordingly (Bethke and Johnson, 2008; Cook et al., 2005; Doyon and Molson, 2012; Engdahl et al., 2013; Green et al., 2014; Maloszewski and Zuber, 1985; Sanford, 1997).

At larger observation scales within given geological units, the widely varying permeability critically enhances spreading and mixing, which translates to an increase of the variability of concentrations (de Dreuzy et al., 2012; Kapoor and Kitanidis, 1998; Le Borgne et al., 2010, 2015) and of the variability of transit times (Cirpka and Kitanidis, 2000; Shapiro and Cvetkovic, 1988). In the

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absence of a general upscaling rule, transit times have been approached using standard solute transport models (Turnadge and Smerdon, 2014), moment-based equations for the mean residence times (Goode, 1996), higher order moments (Varni and Carrera, 1998) or the full probability density function (Cornaton and Perrochet, 2006; Cornaton, 2012) both forward and backward in time (Neupauer and Wilson, 2002). Simulations have further shown how residence time distributions are qualitatively linked to multi-Gaussian permeability structures (Larocque et al., 2009; McCallum et al., 2014a), multi-point geostatistical fields (McCallum et al., 2014b) or stochastic sedimentological structures (Green et al., 2014; Weissmann et al., 2002; Zhang, 2004). While most numerical studies are establishing qualitative relations between heterogeneity and residence times, some have analyzed through inverse problem methodologies the information available in groundwater age data (Ginn et al., 2009; Nassar and Ginn, 2014). Even if inverse problem formalization is standard in groundwater hydrology, it has not been extensively applied because of the additional dispersion and porosity parameters to calibrate (Sanford, 2011).

Residence time variations have been studied up to the formation and regional scales focusing on exchanges and leakages between aquifers (Castro et al., 1998; Sanford et al., 2004; Solomon et al., 2010; Zinn and Konikow, 2007b), and on nested watershed for topography driven flows (Cardenas and Jiang, 2010; Gomez and Wilson, 2013). In addition to the multiple sources of variability within the formation, mixing also occurs within the well or because of wells (Engdahl and Maxwell, 2014; Jurgens et al., 2016; Zinn and Konikow, 2007a), which may collect well-stratified groundwaters with distinct age patterns (Ayraud et al., 2008; Szabo et al., 1996). The impact of sampling is especially important in the convergence zone of flow lines towards adjacent to the system outlets such as streams and wells (Gelhar and Wilson, 1974; Lerner, 1992). Such additional mixing is particularly relevant for field surveys performed on produced wells. Extensive mixing resulting both from the multi-scale transport process within the geological formation and from the sampling conditions would eventually lead to simple broad residence time distributions close to exponential and truncated exponential shapes (Haitjema, 1995; Luther and Haitjema, 1998) traducing complete mixing conditions also found in reactor and blender theory (Danckwerts, 1953).

The residence time distribution may thus be approached by simple models whether they are Dirac distributions for piston flow conditions, inverse Gaussian for homogeneous advective-dispersive conditions, exponential distribution for complete mixing or even composed distributions and more advanced functions depending on the flow conditions. This idea of approaching the actual residence time distribution by a simple model has been formalized as the Lumped Parameter Model (LPM) approach (Maloszewski and Zuber, 1982, 1993). The LPM should appropriately traduce the main flow conditions and be parsimonious in order to be easily calibrated on existing tracer concentration data. Close to a Bayesian spirit, the LPM equilibrates the a priori knowledge on the flow structure traduced by the choice of the distribution function and the quality of the calibration generally taken as the likelihood factor. Interpretation of tracer tests can also be effectively performed within a formal Bayesian framework with appropriate weighting of the model complexity (Massoudieh et al., 2012, 2014). The LPM approach has become standard in the interpretation of atmospheric tracer concentrations. It has been especially used when concentrations of multiple tracers are available and may lead to incompatible groundwater ages when independently interpreted with piston-flow models (Alvarado et al., 2007; Han et al., 2012; Kralik et al., 2014; Morgenstern et al., 2015; Solomon et al., 2010). In such cases (generally outside recharge zones) and when the quantity and diversity of hydraulic and tracer concentrations have allowed some field assessment, the calibrated LPM has been found to have rather wide distribution with coefficient of variations (standard deviation over mean) of the order of one (Blavoux et al., 2013; Gilmore et al., 2016; Green et al., 2014; Kolbe et al., 2016; Leray et al., 2012; Marçais et al., 2015; Massoudieh et al., 2012, 2014). LPMs provide thus important tools for the consistent interpretation of multiple atmospheric tracer concentrations. The approach has been further complemented with additional analytical and empirical distribution models for more complex flow systems (Cvetkovic, 2011; Leray et al., 2016; Zhang et al., 2013). While extensively used, the LPM approach has been assessed only in the few cases where the residence time distributions has been simulated by calibrated numerical models.

In fact, the residence time distribution cannot be measured directly. While the availability of a sufficient number of distinct tracers can in theory allow inverse characterization of the RTD (Massoudieh and Ginn, 2011), such data are typically unavailable, and consequently the RTD can at best be derived from calibrated numerical models. A first direct analysis of residence time distributions obtained from four aquifers in different geological contexts has shown that residence time distributions can be approached by simple analytical distributions even in complex interrelated aquifer systems (Eberts et al., 2012). The a priori choice of the relevant LPM remains however an open issue in complex cases. Further assessments have consisted so far in building calibrated flow and transport models on very few aquifers with residence times ranging over some decades, and in comparing their predictions to those obtained from a priori chosen LPMs calibrated on the same tracer data (Green et al., 2014; Leray et al., 2012; Marçais et al., 2015). LPM predictions were found accurate (within 10%) with well-chosen distribution models. These conclusions remain however very partial and limited to the cases investigated.

Whether it is inferred by use of multiple tracers, calibrated numerical models or as LPMs, the residence time distribution is often the relevant intermediary between the physical and chemical observations and the targeted prediction (Wachniew et al., 2016). Residence times have been extensively used for recharge protection zone delineation (Molson and Frind, 2012), and for estimating contaminant transport and degradation in cases where time can be used as a proxy for reactivity (Bohlke, 2002; Broers and van der Grift, 2004; Pinay et al., 2015). Punctual groundwater age observation might also be combined with groundwater flow and transport models to obtain spatially distributed predictions (Basu et al., 2012; Marçais et al., 2015). More regular use of atmospheric tracers might be further promoted by quick progresses on cost-effective and accurate chemical analytical capacities (Aquilina et al., 2014).

2. Contributions (this issue)

Contributions in this special issue cover a wide range of topics on the origin of RTDs, on their simplification as LPMs, on their identification, and on their application for reactive transport.

Five out of the fourteen articles are dedicated to the characterization of RTDs from varying combinations of field data and simulations over temporal scales ranging from days to millennia. Peralta-Tapia et al. (2016) show with a 10-year isotopic time series ($\delta^{18}0$ and δ^2H) that shallow subsurface flows on a 500-m scale Swedish snow-covered Boreal catchment issue gamma distributed RTDs. Hydroclimatic variability induces significant inter-annual evolution of the derived mean residence time from 300 days to 1200 days linked to the annual rainfall rather than to the snowmelt. Ameli et al. (2016) perform semi-analytical flow and transport simulations based on the configuration of another 100-m scale Swedish catchment with exponentially decaying