Important progress on the use of isotope techniques and methods in catchment hydrology

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Received: 20 January 2009    Accepted: 3 May 2009

ABSTRACT

The use of isotope techniques and methods in catchment hydrology in the last 50 years has generated two major types of progress: (1) Assessment of the temporal variations of the major stocks and flows of water in catchments, from which the estimation of water residence times is introduced in this paper. (2) Assessment of catchment hydrologic processes, in which the interactions between different waters, hydrographical separation, and bio-geochemical process are described by using isotopes tracers. Future progress on isotope techniques and methods in hydrology is toward the understanding of the hydrological process in large river basins. Much potential also waits realization in terms of how isotope information may be used to calibrate and test distributed rainfall-runoff models and regarding aid in the quantification of sustainable water resources management.

Keywords: isotope; hydrological process; catchment; progress

1. Introduction

In 1950s, isotope techniques began to be used to solve a variety of hydrologic and hydrogeologic problems (Li and Zhang, 1990). With the development of isotopic analysis techniques, by studying the waterbodies themselves and some of the isotopic composition of dissolved solutes, isotope techniques and methods have become the modern scientific means of water research (Wei, 1992). Isotope techniques and methods can trace the water cycle effectively—such as indicating water sources, water pathways, and water amount (including rivers and lakes); determining groundwater age; and recording water-rock interactions. Also, isotope techniques and methods can be used to understand the formation and evolution mechanism of water, to provide an important basis for the rational use of water resources (Zhang et al., 2006).

Isotope methods were introduced into catchment hydrology research in the 1960s as complementary tools to conventional hydrologic methods for addressing questions of where water goes when it rains, what pathways it takes to the stream, and how long water resides in the catchment (McDonnell, 2003). Despite slow incorporation into routine research applications, the last decade has seen a rapid increase in isotope-based catchment studies (Vitvar et al., 2005). The progress of isotope techniques in catchment hydrology in recent decades has generated two major sets of applications: assessment of the temporal variations of the major stocks and flows of water in catchments between events such as estimation of water residence times and quantification of recharge travel times; and assessment of catchment hydrologic processes such as quantification of the sources of runoff and delineation of infiltration and exfiltration zones along streams. These have been mainly carried
out in small well-instrumented experimental catchments, on the order of 0.01 to 100 km², and located typically in headwater areas (Buttle, 1998; McGuire and McDonnell, 2006). In contrast, little has been done in terms of application and transfer of these concepts and methodologies to large (>100 to 1,000 km²), less-instrumented experiments in basins (Vitvar et al., 2005). Recently, the International Atomic Energy Agency (IAEA) launched the "large-scale hydrological isotope tracing process" projects, including the arctic, temperate, tropical, arid, lowland, and mountain valley areas. The drainage area and runoff of those areas take up 22 percent and 33 percent of the global land area and global river runoff, respectively.

In the basin scale, studies on isotope hydrology were conducted in the Yellow River Basin (Wang et al., 2004), northern China (Chen et al., 2002; Song et al., 2006), some arid areas (Li et al., 1995; Zhang et al., 2005), and the southern mountainous areas in China (Yin et al., 2000). For various reasons, however, the development and systemic research in China on isotope hydrology are far behind that of developed countries (Song and Yu, 2004). China has a vast territory. The hydrological, geographical, geological, and meteorological factors vary sharply from one place to another. Because of the vast areas and weak infrastructure, it is very difficult to carry out research work on catchment hydrology with the traditional methods. Therefore, use of isotopes and hydrogeochemical techniques and methods to promote the understanding of the hydrological cycle and water resources research and management and to achieve sustainable development of the basins is essential. In this paper, we review the major applications of isotopes to catchment studies and address a variety of prospective new directions in research and practice.

2. The transit time of water in a catchment

The hydrologic cycle in catchments varies in time (Ohmura and Wild, 2002). Water infiltrates at different rates, mixes in the subsurface, and thus has different travel times to the stream. In fact, streamwater is an integrated mixture of water with various sources or different ages (or residence time) that contributes to streamflow generation that fell on the catchment in the past. The transit time through the stream network (e.g., hyporheic and channel) is generally much shorter than transport through the catchment’s subsurface and thus has generally been ignored in catchment transit time distributions (Kirchner et al., 2000; 2001). The transit time of water in a catchment is the average time elapsed after a water drop entered the catchment and the time it is observed in the catchment outlet, well, or soil depth (Vitvar et al., 2005).

The time water spends traveling subsurface through a catchment to the stream network (i.e., the subsurface transit time) is a fundamental catchment descriptor that reveals information about the storage, flow pathways, and source of water in a single characteristic. Transit time is directly related to internal catchment processes. The distribution of transit times describes how catchments retain and release water and solutes that, in turn, control geochemical and biogeochemical cycling and contamination persistence (Burns et al., 2003). Longer transit times indicate greater contact time and subsurface storage, implying more time for biogeochemical reactions to occur as rainfall inputs are transported through catchments toward the stream channel. Thus, quantifying the mean residence time provides a primary description of the hydrobiogeochemical system (Wolock et al., 1997), catchment sensitivity to anthropogenic inputs (Turner et al., 2006), and land-use change (Burns et al., 2005).

2.1. Isotopes for estimating transit time in catchment hydrology

Seasonal variations of stable isotopes can be used to estimate the transit time of groundwater. Stable isotopes deuterium (D) and oxygen (18O) are part of water molecules themselves. These tracers are applied by precipitation and are generally distinct isotopically, which makes them reliable tracers. Because of the conservative nature of those isotopes and the ease in field and laboratory processing of water samples, D and 18O have been the dominant age-assessment tools. The amplitude of seasonal variations in δ18O is attenuated during groundwater recharge, and the preservation of seasonal variations signifies short residence time or flowpath. Generally, D and 18O are for dating waters up to about five years of age, which typically occur in shallow aquifers connected to streams (Maloszewski and Zuber, 1982; Amin and Campana, 1996; Ozyurt and Bayari, 2005; McGuire and McDonnell, 2006).

Based on the half-life of radioactive decay, the transit time of groundwater can be inferred from radioisotopes. Natural 14C (carbon) was discovered in the late 1940s, and natural 3H (tritium) was discovered in the early 1950s. Shorty thereafter, atmospheric nuclear-weapon tests substantially increased the 3H content in the rapidly circulated parts of the hydrologic cycle, with the peak at around 1963. Tritium was therefore used for the first systematic estimations of water age in catchments (Eriksson, 1958). The content of 3H in groundwater decayed by rocks is tiny and can be neglected. If 3H in groundwater is detected, it means the groundwater is newly recharged. A larger than 30TU 3H content indicates the groundwater was recharged in 1960s. Now, 3H is rarely used for water age-dating. 14C, which has a much longer half-life (5,730±40 yr), is a common and effective radio isotope to date the ancient or deep groundwater (Wassenaar et al., 1991).

Noble gases, such as 3He (Torgersen et al., 1979; Solomon et al., 1993), 85Kr (Smethie et al., 1992), and 222Rn (Rogers, 1958) have been used to estimate mean residence time. While environmental tracers or anthropogenic compounds—namely 35S (Lal and Peters, 1966), Be (Cooper et al., 1991), chlorofluorocarbons (CFCs) (Thompson et al., 1974; Plummer and Busenberg, 2000), sulfur hexafluoride...
(SF$_6$) (Maiss and Levin, 1994)—can be used to date water ages from days to decades. These tracers are not applicable to surfacewaters because of contamination by exchange with atmospheric and vadose zone gases (Plummer and Busen-berg, 2000).

Two or more isotopes can be used at the same time to estimate mean residence time. The results of mean residence time, which have been gotten by different isotopes, can be contrasted and verified (Ekwarzel et al., 1994; Plummer et al., 2001; Zhang et al., 2005).

2.2. Transit time modeling

There are many modeling approaches to estimate transit times. These include compartment models (Campana and Simpson, 1984), conceptual hydrologic models (Lindström and Rodhe, 1986), stochastic–mechanistic methods (Destouni and Graham, 1995; Simic and Destouni, 1999), particle tracking (Molénat and Gascuel-Odoux, 2002), and direct simulation (Goode, 1996). Often, these approaches require hydrological characterization of the catchment in developing models to approximate transit times. Many catchments lack data to benefit from these techniques.

The lumped-parameter approach is widely used to infer transit times from tracer data (natural or applied). Lumped-parameter methods provide estimates of catchment-scale hydrological parameters (i.e., mean transit time, transport velocities, storage) through an inverse procedure where the parameters of a transit time distribution (TTD) are estimated by calibrating simulations to fit measured tracer output composition (Maloszewski and Zuber, 1982; Zuber, 1986). Mathematically, the transport of conservative tracer through a catchment can be expressed by the convolution integral, which states that the stream outflow composition at any time—\( \delta_{\text{out}}(t) \)—consists of tracer, \( \delta_{\text{in}}(t-\tau) \), that fell uniformly on the catchment in the past \((t-\tau)\), which becomes lagged according to its transit time distribution, \( g(\tau) \):

\[
\delta_{\text{out}}(t) = \int_{0}^{\infty} g(\tau) \delta_{\text{in}}(t-\tau) d\tau = g(t) * \delta_{\text{in}}(t) \tag{1}
\]

where \( \tau \) are the lag times between input and output tracer composition, and the asterisk represents the shorthand of the convolution operation. Figure 1 illustrates the lumped-parameter-model concept for determining the transit time of water draining a catchment. Environmental tracers are applied naturally during precipitation (e.g., $^{18}$O) and are transported to the stream network along diverse surface and subsurface flowpaths within the catchment. In most undisturbed catchments, however, flowpaths are predominantly subsurface. The transport process along subsurface flowpaths causes delay (due to advection) and spreading (dispersion) of tracer arrival in the stream network, which is a direct reflection of the catchment’s flowpath distribution, runoff processes, and subsurface hydrologic characteristics. The result of differential transport within the catchment is a tracer output signal (baseflow) that is damped (i.e., decreased in standard deviation and amplitude and lagged compared to the input signal). The complex distribution of catchment flowpaths is represented by a distribution of transit times, \( g(\tau) \), which describe the integrated behavior of tracer transport through the catchment. The integrated response of tracer arrival at the catchment outlet from all locations in the catchment is described by the TTD (transit time distribution).

Models for transit time distribution (TTD) include Piston Flow Model (PFM), Exponential Flow Model (EM), Combined Exponential-Piston Flow Model (EPM), Dispersion Model (DM), Linear Model (LM), and Combined Linear-Piston Flow Model (LPM). It is difficult to choose an appropriate TTD model because of the short series of isotope data in practice. The choice and assessment of the TTD models are presented in Maloszewski and Zuber (1982); Zuber (1986); Maloszewski and Zuber (1996); and McGuire and McDonnell (2006).

These techniques on the lumped-parameter transit-time-modeling approach have been recently formalized into a variety of software packages such as MULTIS (Rich-ter et al., 1993), FLOWPC (Maloszewski and Zuber, 1998), TRACER (Bayari, 2002), LUMPED (Ozyurt and Bayari, 2003), and LUMPED unsteady (Ozyurt and Bayari, 2005).

The lumped-parameter approaches of estimating transit times for streams and catchments provide a quantitative approach to fundamentally describing the catchment flow system. The approach relies primarily on tracer data and, thus, is useful in gauged and ungauged basins and as a complement to other types of hydrological investigations. A critical analysis (McGuire and McDonnell, 2006) of unresolved issues should be evaluated in future research through the application of lumped-parameter transit-time modeling at the catchment-scale. These issues included (1) the input characterization issue, (2) the recharge assumption, (3) the data record length problem, (4) the stream-sampling issue, (5) the transit-time distribution-selection problem, and (6) the model-evaluation process.

2.3. Factors that influence transit time

The research on transit time has been mainly in small basins. Recently, some studies have been carried on in larger areas (Aggarwal, 2002; Gibson et al., 2002). The relationship between basin area and baseflow residence time remains equivocal (Vitvar et al., 2005). Though no studies have reported a relation between residence time and catchment size, McDonnell et al. (1999) and McGlynn et al. (2003) found that the internal flowpath composition may be a first-order control on stream baseflow age. Recent $^{18}$O and $^3$H studies comparing small catchments in Japan and New Zealand show how bedrock permeability may control the direction of water aging (Uchida et al., 2004). In the impermeable bedrock case, Stewart and McDonnell (1991)
observed a lateral downslope increase in soil water mean residence time. Asano et al. (2002) tested this hypothesis on a comparable slope configuration, but with permeable bedrock, and found that water aged vertically through the soil profile, with no evidence of a downslope age increase. In this case, the communication of water vertically between the soil and underlying bedrock did not “force” a downslope component to soil water age.

![Conceptual diagram of the lumped-parameter transit-time-modeling approach (after McGuire and McDonnell, 2006)](image)

3. Hydrological processes assessment in catchment

As we have mentioned previously, different sources of water have different ages. Regardless of the age of streamwater, runoff in streams is generated from a variety of spatial sources and along various flow pathways. This complexity increases with catchment size, so that large rivers often represent highly heterogeneous mixtures of water types. Isotopic and geochemical tracers are sensitive to the physical and chemical behavior of natural elements and compounds (Gibson et al., 2005). The variations of isotopes and geochemical compositions supply the necessary information on the hydrological process in a basin.

3.1. The interaction between streamwater and groundwater

For the study of interaction between streamwater and groundwater, Darcy’s law, accompanied by the regional hydrogeological conditions and some hydrogeological parameters, is mostly used in traditional methods. However, traditional methods are restricted in complicated spatial interaction between surfacewater and groundwater (especially in areas with complex hydrogeological conditions). The differences of spatial distribution of isotopic composition in water can be used to reveal the interaction between waterbodies.

The concentration of stable isotopes, D or $^{18}$O, is mainly influenced by the mixture and physical conditions such as evaporation and condensation, which causes isotope fractionation. Evaporation effects lead to the enrichment of heavy isotopes in lakes, rivers, drainage channels, and shallow groundwater (Simpson and Herczeg, 1991) and makes water line deviations from global meteoric water line (GMWL) or local meteoric water line (LMWL). The deviation is particularly evident in arid and semi-arid regions (Friedman et al., 1992; Zhang et al., 2006). By using such differences, McCarthy et al. (1992) studied the interaction...
between riverwater and groundwater of the Columbia River, as it flows through Portland, Oregon. Using the isotopic and chemical data of springs, streamwater, and groundwater along the river channel, Song et al. (2006) studied spatial distribution and evolution trends of isotopes and chemical composition in the Huaisha River Basin, determined the sources of surface water and groundwater and the flow path, and came to a conclusion that the basin is non-closed (i.e., also accepts water from outside of the basin). Literature on such research is numerous and not listed in this paper.

The differences of environmental isotopic contents of water in different aquifer systems in complicated hydrogeologic regions reveal complicated hydraulic connections between waters. Using isotope methods in such complicated regions is convenient, fast, and has economic advantages. Using 34S as a tracer, Rightmire et al. (1974) found the groundwater in a limestone aquifer in Texas was recharged by unconfined groundwater. Based on the isotopic data of 18O and 2H, Yurtsever and Payne (1974) analyzed the salty water recharge from a deep, confined aquifer to unconfined groundwater in southwestern Qatar. Incorporating additional relations between 38O and Cl, a conclusion was drawn that unconfined groundwater was recharged by deep confined groundwater, seawater, and local precipitation.

Because of contamination by exchange with atmospheric and vadose zone gases, noble gases isotopes are mainly used in groundwater study at present. However, Schlosser et al. (1988) introduced a method of calculating streambed infiltration velocities using the 3H/3He ratio. These techniques have been suitable in areas of both high and low recharge rates. In larger drainage basins, the process of infiltration from rivers into river banks can be successfully addressed using isotopic approaches. Schlosser et al. have quantified this connection using 3H/3He. 222Rn could be used for studying leakage of river water into shallow aquifers (Ellins et al., 1990) and delineation of exfiltration and infiltration zones along river reaches (Wu et al., 2004). In zones where river baseflow exfiltrates into the adjacent aquifers, recharge velocities and residence times of the recharged water can be obtained by use of He techniques (Solomon et al., 1993). Conversely, the source and residence times of groundwater seepage to streams have been evaluated by using chlorofluorocarbons (Modica et al., 1998).

3.2. Hydrograph separation

Rainfall-runoff is a key part of the hydrological cycle. The main contents of a rainfall-runoff study are the distribution of runoff discharge and rainfall-runoff unit hydrograph. In the 1950s, scholars tried to use the salinities dissolved in precipitation to study the formation of the surface-runoff mechanism (Zhang et al., 2006). Using variations of chemical contents in precipitation and streamwater to study the rainfall-runoff process was much improved by the 1970s. Along with the application of geochemistry, isotopic techniques were gradually applied to the rainfall-runoff process (Kendall and Coplen, 2001).

Since the late 1960s, D and 18O have been used routinely to delineate the baseflow component of a stormflow event, where event water is represented by the distinct isotopic composition of rainfall or throughfall, and pre-event water is represented by the distinct isotopic composition of pre-storm streamwater or adjacent groundwater. This approach has been applied in a large number of studies of stormflow events in small catchments (Pearce et al., 1986; Turner and Barnes, 1998). Because of the complexity of hydrological system and runoff-formation process, some basic assumptions are often contained in hydrograph separation. Buttle (1994) commented on the reliabilities of those assumptions.

In general, these studies on hydrograph separation have revealed a much greater baseflow proportion in the stream discharge hydrograph, which differs markedly from the early conceptual models of streamflow generation and graphical hydrograph-separation analysis (Fritz et al., 1976; McDonnell et al., 1991). A study of statistics on hydrograph-separation studies in Canada by Gibon et al. (2005) showed that more than 60 percent of the runoff was from pre-event water stored in river basins, especially in the vast majority of forest-covered basins or wetlands watershed. By using 1H and 18O, Gu et al. (2003) studied the relation between rainfall and runoff in an experimental area in Chuzhou, China. The results showed that it was incorrect that the surface runoff must be derived from the event precipitation. In some cases the pre-event water accounts for more than half. Kirchner (2003) supplied theoretical discussions on those phenomena.

The joint application of isotope tracers and geochemical tracers (such as trace elements, major ions, dissolved organic carbon, etc.) makes it possible to identify the interaction between water and bottom layer (e.g., rocks or organic layer). Thus, we can separate a hydrograph into three or more components (Gibon et al., 2005). Application of those methods is seen in Buttle and Peters (1997). Multi-tracer separation methods of runoff sources have been formalized into the concept of End-Member-Mixing-Analysis (EMMA), based on the multivariate statistics and quantification of runoff sources as statistical end-members (Christophersen and Hooper, 1992). EMMA has become a popular technique for interpretation of isotopic (and solute) data and has great potential in extended future applications.

Hydrograph-separation techniques have evolved and become more sophisticated, adopting methods to quantify errors and uncertainties (Joerin et al., 2002). One of the lingering challenges in the application of the runoff tracer-based data is in parameterization and calibration of distributed rainfall-runoff catchment models. Although the isotopic investigations on runoff generation substantially changed the conceptualization of the catchment rainfall-runoff process, they have not been widely incorporated into sophisticated rainfall-runoff models (Beven and Freer, 2001; Leavesley et al., 2002; Gurtz et al., 2003), model
structures, and model parameter testing. Yet, they provide a higher quality of understanding of the runoff components within the model-routing procedure. These approaches are considered as promising means to enhance the relation between isotope-based and conventional hydrologic methods in catchment hydrology (Seibert and McDonnell, 2002).

3.3. Research on biological and geochemical processes

From the point of view of water resources, the quantity and quality of groundwater have equal importance. Solute isotopes such as $^{87}\text{Sr}$ (Stueber et al., 1987), $^{13}\text{C}$ (Spence et al., 2005), $^{34}\text{S}$ (Thomson et al., 1994, and $^{35}\text{N}$ (Clough et al., 2007) provide information on the biological and geological sources of recharge water for groundwater. Thus, isotopes of solutes have become useful tools for research on biological and geochemical processes such as groundwater-recharge mechanisms, groundwater management, groundwater-pollution assessment, land-use effects, nutrients, and water-quality evaluation in catchment. Take $^{15}\text{N}$ as an example.

In recent years, much attention has been paid on the rising trends of NO$_3^-$ pollution in groundwater. Using environmental isotopes, study of groundwater pollution becomes possible. Nitrogen isotopes have been widely used for the study of source identification and reaction mechanisms, and oxygen isotope can be used to effectively identify denitrification (Bottcher et al., 1990). The combination of $^{15}\text{N}$ and $^{18}\text{O}$ of nitrate have been developed and successfully applied on the catchment scale as tracers of nitrate sources in catchments. Bohlke and Denver (1995) showed that, in a small catchment draining into Chesapeake Bay, the outflowing water continued to be contaminated with nitrates for two to three decades after cessation of the N input. A large number of studies also provided information on the contributions of nitrate from precipitation and from microbial nitrification, from microbial denitrification in shallow aquifers, from septic tank leakages, and from animal waste.

By tracing the geochemical reactions in catchment, several other cosmogenic ($^{10}\text{Be}$, $^{18}\text{Be}$, $^{24}\text{Na}$, $^{41}\text{Ca}$) and lithogenic ($^{6}\text{Li}$, $^{37}\text{Cl}$, $^{11}\text{B}$, $^{143}\text{Nd}$, $^{206}\text{Pb}$, $^{207}\text{Pb}$, $^{208}\text{Pb}$, $^{210}\text{Pb}$) isotopes have been introduced into catchment hydrology research within the last two decades. Those isotopes are used to trace the origins and evolution of rivers (Bullen et al., 1994; Brown et al., 1995) and migration of contamination (Vengosh et al., 1994; Bacon et al., 1995). Many potential applications are yet to be realized (Vitvar et al., 2005).

4. Future directions

The application of isotopes to track sources and movement of water in catchments has, over the past 40 years, resulted in a substantial improvement in the understanding of runoff processes. With the development of the isotope techniques and related disciplines, using these techniques and methods to study the catchment hydrology will be much improved and strengthened.

First, with the development and renovation of the means of measuring, more and even new isotopes are likely to be used in the isotope hydrological study. Owing to the ever-decreasing signal of bomb tritium in natural systems, development of new methods for tracing older waters (with mean residence time > 50 yr) is essential. In particular, further investigation and addressing in catchment studies are needed of the use of dissolved gases $^3\text{He}$ and $^{222}\text{Rn}$; solute isotopes $^{15}\text{N}$, $^{34}\text{S}$, $^{87}\text{Sr}$, $^{208}\text{Pb}$, $^{41}\text{Ca}$, $^{11}\text{B}$; and anthropogenic gases CFCs and SF. A movement towards multi-isotope studies is highly desirable; yet, many researchers and subcommunities are still too fixated on one particular isotope. A significant potential also remains in novel applications of isotopes coupled with solutes and artificial tracers in complex approaches such as EMMA or the other geochemical models.

Second, calibration and verification of rainfall-runoff catchment models is an area in which isotopic applications could significantly aid in the process and with parameter interpretation. Through the sampling and analysis of isotopes in atmospheric precipitation, surfacewater, soil water, groundwater, and plant water in a basin, a better understanding of basin precipitation, evapotranspiration, infiltration, and runoff processes can be achieved. Using water as a link, mathematical models on different water interactions are set up to provide a better understanding of hydrological processes in a catchment. With a deep understanding of the hydrological cycle, quantitative, management study of sustainable water resources will come true.

In addition, hydrological scale is one of the hot topics in the area of hydrology research. Recent trends indicate a continually growing interest in isotopic applications to solve practical problems in hydrology and water-resources management in large-scale catchments (Aggarwal, 2002; Gibson et al., 2002; Vitvar et al., 2005). Large basins are very heterogeneous and are typically driven by a large variety of runoff processes. Therefore, the use of methods developed in small natural catchments might be limited in large basins. Techniques and indicators are required to describe the principal processes in large catchments without the need of spatially intensive experimental datasets. Large-scale catchment studies may be able to utilize well-known isotopic effects (the altitude effect, the continental effect, etc.) to assess gross first-order controls on flow generation (Vitvar et al., 2005). Sustainability indicators of water resources are focused typically on local water scarcity. New approaches should be developed to generate, quantify, and integrate indicators of runoff processes in catchments and to evaluate them in terms of potential sustainability.

Acknowledgments:
This research was conducted within the Nation Basic Research Program of China (973 Program, Research No. 2007CB411502) and project "Matter fluxes in Inner Mongolia as influenced by stocking rate (MAGIM)" funded by the German Science Foundation (DFG) (Research Unit 536).
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