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Where does streamwater come from in low-relief forested watersheds? A dual-isotope approach

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Abstract. The time and geographic sources of streamwater in low-relief watersheds are poorly understood. This is partly due to the difficult combination of low runoff coefficients and often damped streamwater isotopic signals precluding traditional hydrograph separation and convolution integral approaches. Here we present a dual-isotope approach involving ^{18}O and ^{2}H of water in a low-angle forested watershed to determine streamwater source components and then build a conceptual model of streamflow generation. We focus on three headwater lowland sub-catchments draining the Savannah River Site in South Carolina, USA. Our results for a 3-year sampling period show that the slopes of the meteoric water lines/evaporation water lines (MWLs/EWLs) of the catchment water sources can be used to extract information on runoff sources in ways not considered before. Our dual-isotope approach was able to identify unique hillslope, riparian and deep groundwater, and streamflow compositions. The streams showed strong evaporative enrichment compared to the local meteoric water line ($\delta^2\mathrm{H} = 7.15 \cdot \delta^{18}\mathrm{O} + 9.28$ ‰) with slopes of 2.52, 2.84, and 2.86. Based on the unique and unambiguous slopes of the EWLs of the different water cycle components and the isotopic time series of the individual components, we were able to show how the riparian zone controls baseflow in this system and how the riparian zone "resets" the stable isotope composition of the observed streams in our low-angle, forested watersheds. Although this approach is limited in terms of quantifying mixing percentages between different end-members, our dual-isotope approach enabled the extraction of hydrologically useful information in a region with little change in individual isotope time series.

1 Introduction

The spatial and temporal sources of runoff in low-angle, forested headwater watersheds are poorly understood. Most of what we know of runoff generation in forested terrain comes from steep humid sites where elevation potential dominates and runoff responses are high (for a review, see Bachmair and Weiler, 2011). Much recent work has focused on the threshold sequencing of spatial sources in upland forested watersheds (Sidle et al., 2000; Seibert and McDonnell, 2002), hillslope–riparian connectivity (McGlynn and McDonnell, 2003), and the importance of spatial patterns of hillslope–riparian–stream connectivity (Jencso et al., 2009; Jencso and McGlynn, 2011). Such connectivity may be strongly nonlinear (Buttle et al., 2004; Zehe et al., 2007; Penna et al., 2011). Consequently, streamflow chemistry in upland forested watersheds is often determined by volume ratios of water sourcing in the hillslopes compared to riparian-zone water (McGlynn and McDonnell, 2003), with

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many watersheds showing only brief expressions of adjacent hillslope water chemistry during large rainfall and snowmelt events (Burns et al., 2001).

Unlike the distinct watershed components found in steeper headwater counterparts (hillslope, hollow, riparian), low-angled terrain blurs the boundary between the riparian zone and hillslope and presents little in the way of obvious geomorphic units that might be considered for model construction. Early work in low-angled terrain showed how matric potential (rather than elevational potential) dominates total potential and the resulting subsurface runoff flow paths (Anderson and Kneale, 1982). More recent work in lowland forests has shown that runoff may be generated from only small proportions of the watershed (Devito et al., 2005a). Lowland areas often exhibit a complex groundwater–surface-water interaction. Water fluxes between slopes and wetlands are generally small (Devito et al., 2005a; Branfireun and Roulet, 1998), and hillslope–stream connectivity is rare (Redding and Devito, 2010; Ali et al., 2011). These features in lowland forested watersheds appear to be controlled by the complex, and poorly understood, interplay of climate, soils, and geology (Devito et al., 1996, 2005b; Slattery et al., 2006; Sun et al., 2002). Furthermore, topography is not a clear driver of runoff generation (Buttle et al., 2004; Devito et al., 2005b) since vertical subsurface flow often dominates over lateral subsurface flow (Todd et al., 2006). Saturation excess overland flow often dominates the runoff response in these areas (Eshleman et al., 1994; Slattery et al., 2006; La Torre Torres et al., 2011), but the linkages between hillslopes, riparian zones, and the stream are difficult to observe, conceptualize, and quantify.

Ordinarily, streamwater stable isotope tracing and isotope hydrograph separation would help with questions of source components of streamflow (Klaus and McDonnell, 2013). However, areas with low runoff coefficients, small event water contributions, or long transit times have stream isotopic signals that are difficult to decipher due to the damping of the atmospheric input signal. Despite this, La Torre Torres et al. (2011) have noted the pressing need for isotope studies to "identify the sources of storm flow and base flow to better understand flow generation mechanisms" in watersheds in low-relief areas (in their case, the Atlantic Coastal Plain of the USA).

So what can be done in low-relief areas to quantify runoff sources when streamwater isotope signals are muted and the flow itself in headwater streams is often very ephemeral? Here, we present new work that addresses this fundamental question using a dual-isotope approach involving ^{18}O and ^{2}H. While numerous studies have used water lines based on dual isotopes in various water cycle applications (Gonfiantini, 1986; Gibson et al., 2008, 2010; Yi et al., 2010; Zhang et al., 2013), we are unaware of any to date that have used this approach to determine streamwater source components and, hence, use them to build a conceptual model of streamwater generation. We concentrate our efforts here on the lowland forested watersheds draining the Savannah River Site (SRS) in the Coastal Plain of South Carolina, USA, and show the relationships of ^{2}H and ^{18}O for various water cycle components in three headwater catchments over a 3-year observational period. We show proof of concept of this approach to quantify the source(s) of streamflow, particularly during baseflow conditions. We present evidence that the slopes of the meteoric water lines/evaporation water lines (MWLs/EWLs) of the catchment water sources may be used to extract information on runoff sources in ways not considered before. We then show how these distinct slopes may be an aid to separate and quantify where streamwater comes from in our low-angle, forested watersheds and develop a conceptual understanding of where water comes from in these catchments. Lastly, we use a combination of δ^{18}O and δ^{15}N of nitrate to compare to our dual-isotope interpretation of water contributions to streamflow.

2 Study site and methods

2.1 Study area

The study was conducted in three adjacent forest headwater watersheds that are tributaries to Upper Fourmile Branch, at the Savannah River Site, a National Environmental Research Park. The three watersheds have areas of 0.45 km^2 (R watershed), 1.69 km^2 (B watershed), and 1.17 km^2 (C watershed). The watersheds are located within the Aiken Plateau of the Upper Atlantic Coastal Plain in South Carolina, USA (Fig. 1). Average annual precipitation is 1225 mm distributed evenly throughout the year (Fig. 2). The climate is characterized by long, hot summers with an average daily maximum temperature of 32.3 °C and relatively mild winters with an average temperature of 8.6 °C (Rebel, 2004). The measured average annual pan evaporation over 30 years was 1448 mm (Blacksville, SC, ∼25 km distance from SRS) (Kilgo and Blake, 2005) and the calculated average annual potential evapotranspiration is 1443 mm, based on the Priestley–Taylor equation (Rebel, 2004). Actual evapotranspiration is approximately 90 % of the potential (Riha and Rebel, 2004; Samuelson et al., 2006). Potential transpiration is about 95 % of potential evapotranspiration in the summer and 82 % in the winter (Rebel, 2004). On six experimental plots throughfall was reduced by 10.1 to 16.4 % compared to open precipitation (Hitchcock and Blake, 2003). Annual runoff coefficients are as low as 0.01 (Du et al., 2014). The R watershed ranges from 70 to 106 m a.s.l. (meters above sea level), the B watershed from 80 to 108 m a.s.l., and the C watershed from 70 to 103 m a.s.l. The upslope areas are characterized by gently rolling hills with an average slope of ∼2–3 %; stream valleys (representing the riparian zone) consisted of long, flat, forested wetlands as well as Carolina Bay wetlands that are characteristic of the Upper Atlantic Coastal Plain. The hillslopes and ridges are covered by longleaf

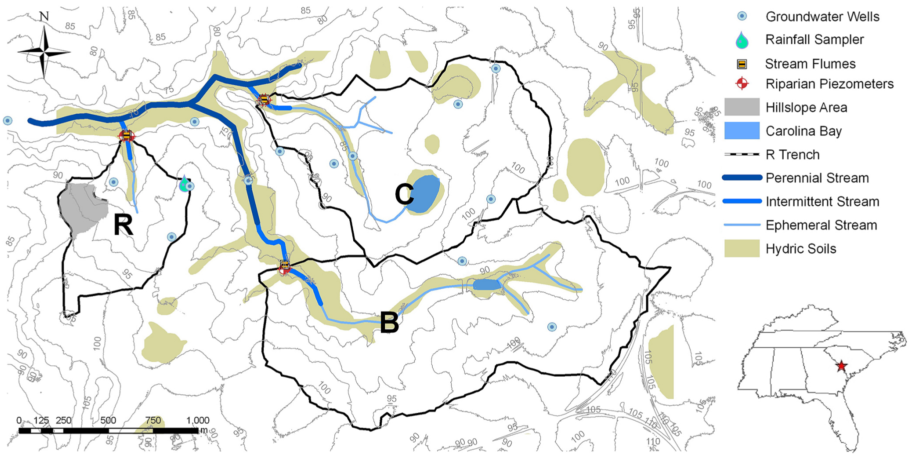

Figure 1. Study site with the three watersheds (R, B, C), the trenched hillslope, streams, instrumentation, the distribution of hydric soils, and the location within the US.

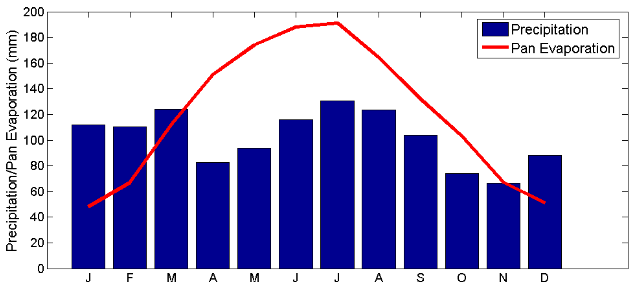

Figure 2. Average monthly precipitation and monthly pan evaporation for the study area (data from Kilgo and Blake, 2005).

pine (*Pinus palustris*), loblolly pine (*P. taeda*), and slash pine (*P. elliottii*), while mixed hardwoods, mainly sweet gum (*Liquidambar styraciflua*), dominate the riparian areas. The soils are well-drained, loamy, siliceous, thermic Grossarenic Paleudults (Rasmussen and Mote, 2007), with an argillic Bt horizon. Hydric soils occupy the riparian zone and depressions such as wetlands and Carolina Bays. Surface soils contain 80–90 % sand; the clay content increases to 35 % or more in the Bt horizon (Kilgo and Blake, 2005). In situ hydraulic conductivity (K_{sat}) measurements with a compact constant head permeameter indicate medians around 10 cm h^{-1} in the topsoil and 0.5 cm h^{-1} in the argillic horizon, with anomalies of clearly higher K_{sat} (Du et al., 2014) allowing vertical recharge. Mapping of the depth to the argillic horizon in a 40 m $\times$ 40 m plot (2 $\times$ 1 m grid) in the R watershed revealed an average depth of 0.76 m (ranging from 0.19 to 1.62 m). At three excavated trenches (30–121 m), the depth to clay showed median values of 0.5–0.8 m and ranged from 0.15 to 2.0 m, and the thickness of the argillic layer varied from 1.3 to 3.0 m, with a mean thickness of 2.1 m (Du et al., 2014). From bottom to top, the underlying geology consists of Late Cretaceous quartz sand, pebbly sand, kaolinitic clay, Paleocene clayey and silty quartz sand, glauconitic sand, and silt (Wyatt and Harris, 2004).

2.2 Sampling and isotope analysis

Sampling at the site is an ongoing process. In this paper we chose to limit the data to that collected until mid May 2012 (records started in mid 2010 in watersheds B and C and 2007 for watershed R), as a harvest of 40 % of the forests in watersheds B and C was performed in spring 2012 and completed by May 2012.

At the outlet of each watershed, an H flume and automatic sampler (ISCO 6712, Teledyne ISCO, Lincoln, NE) were installed to collect streamwater samples and record water level for calculation of streamflow. Sampling of streamwater was done by the automated sampler and grab samples on a weekly basis. The R stream was sampled from April 2007 and the B and C streams from March 2010 until the streams fell dry during May 2011. Adjacent to each stream gauge (Fig. 1), two shallow piezometers were installed in the riparian zone to sample riparian groundwater from the hydric soil at monthly intervals. Event-based (six events between February 2011 and May 2012) lateral subsurface flow was sampled at a 120 m trenched hillslope (0.057 km^2) in the R watershed, either as composite samples for events or with several discrete samples per event. Precipitation was sampled at approximatley weekly intervals with a bulk sample (February 2007 until May 2012). Evaporation-influenced samples were removed from the data set (deuterium excess < 0 and precipitation amount < 3 mm). Throughfall was sampled weekly to biweekly at three locations within each catchment (starting November 2010), where a ~ 200 cm^2 funnel collected the water. Groundwater was sampled from 14 wells, all located in the same strata, between 2 and 12 times per well over an 8-month period from September 2011 to May 2012. The water samples were analyzed for stable isotopes of water; the ratio

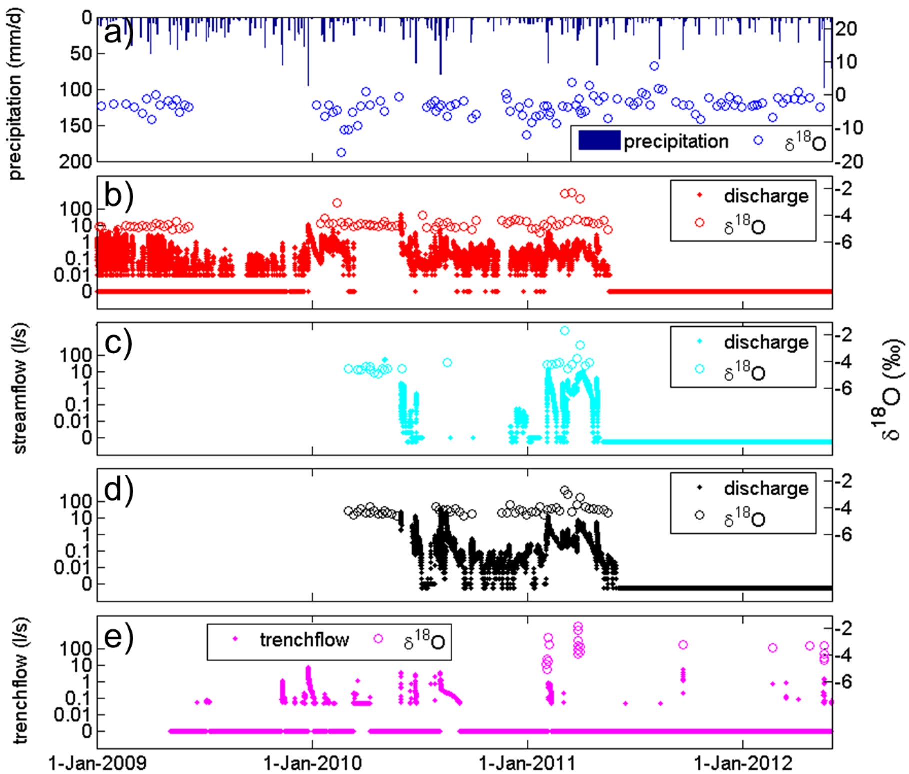

Figure 3. (a) Daily precipitation amount and δ^{18}O of precipitation; **(b)** streamflow and δ^{18}O in the R watershed; **(c)** streamflow and δ^{18}O in the B watershed; **(d)** streamflow and δ^{18}O in the C watershed; **(e)** trenchflow and δ^{18}O in the hillslope trench of the R catchment.

of ^{2}H/^{1}H and ^{18}O/^{16}O of liquid water samples was measured with a Los Gatos Research (LGR) liquid water isotope analyzer (LWIA) that utilizes off-axis integrated cavity output spectroscopy (Baer et al., 2002) and was converted to δ^2H and δ^{18}O using the VSMOW.

For stable isotopes of nitrate, water samples were collected in the field from streamwater, riparian groundwater, throughfall, and lateral flow from the trenches. Water samples were immediately filtered (GF/F, Whatman Inc.) into acid-washed, HDPE bottles and frozen until analysis. Nitrate concentrations were measured using the cadmium reduction method (APHA 2005) on a SEAL Analytical AA3 autoanalyzer. Stable isotopes of nitrate were measured using the denitrifier method with *Pseudomonas aureofaciens* and *P. chlororaphis* bacteria (Sigman et al., 2001; Casciotti et al., 2002) at the UC (University of California) Davis Stable Isotope Facility. The ratios of ^{15}N/^{14}N and ^{18}O/^{16}O were measured on a Thermo Finnigan Gas Bench and PreCon trace gas concentration system with a Thermo Scientific Delta V Plus isotope-ratio mass spectrometer, and a minimum of 1 µM NO_3 was required for analysis. $\delta^{15}N_{NO_3}$ and $\delta^{18}O_{NO_3}$ were determined against standards USGS 32, USGS 34, and USGS 35 and were reported relative to N_2 in air for $\delta^{15}N_{NO_3}$ and relative to the Vienna Standard Mean Ocean Water (VSMOW) for $\delta^{18}O_{NO_3}$.

3 Results

3.1 Hydrological and isotopic dynamics

Precipitation totaled 1373 mm in 2009, 964 mm in 2010, and 989 mm in 2011 (Fig. 3a). The below-average annual precipitation amount in 2010 and 2011 led to dry streams from spring 2011 until the end of the observation period. Generally, streamflow in all three streams was intermittent with zero-flow periods. Streamflow was usually generated when the wetland zone in the valley bottom was saturated. Some of the precipitation (Fig. 3a) events generated short-lived hydrograph peaks in the three watersheds and the hillslope trench (Fig. 3b–e). Overall, storm runoff ratios were extremely low (< 2.3 %) and streams, even when flowing, were very muted in the response to heavy rainfall (Fig. 3b–d). While some deeper groundwater wells showed groundwater depths of ~ 10 m, the well in the riparian zone of the C watershed approached the soil surface (< 1 m) during wet periods.

The isotopic ratios in precipitation varied between -17.3 and $+3.9$ ‰ for ^{18}O and -122.7 and $+37.4$ ‰ for ^{2}H (Figs. 3a and 4a). The δ^{18}O and δ^2H values for streamwater were much less variable, and averaged around -4 and -23 ‰, respectively, for all three streams. The streamwater values varied between -5.5 and -2.3 ‰ (^{18}O) and -26.6 and -18.0 ‰ (^{2}H) for the R stream, -4.9 and

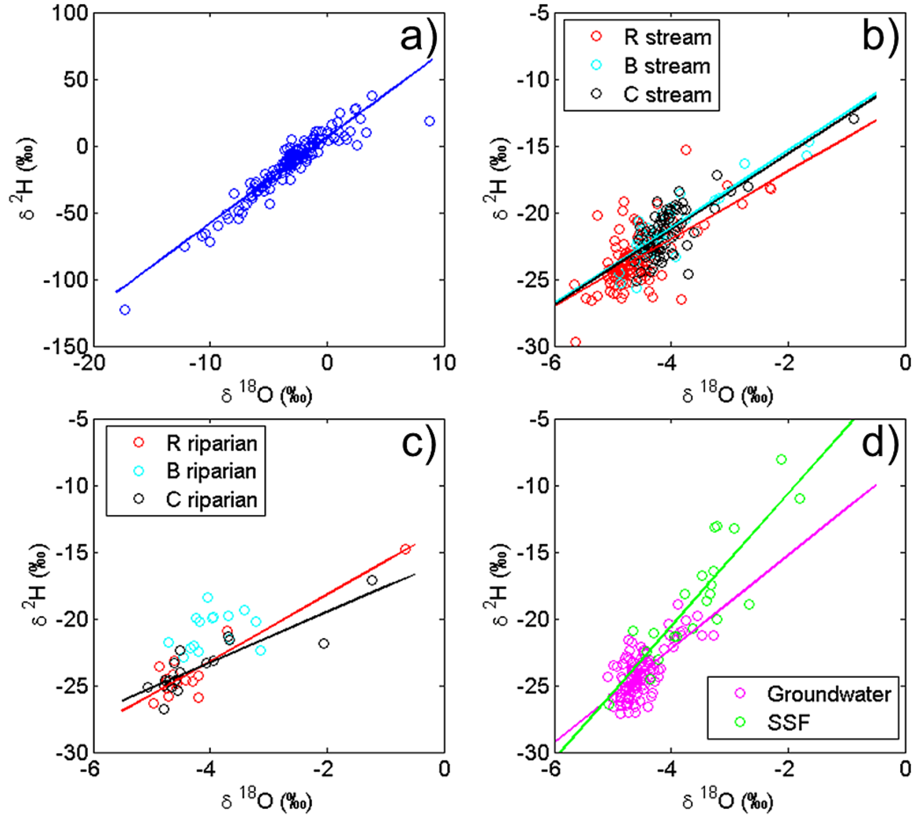

Figure 4. ^{2}H versus ^{18}O and the meteoric and evaporation water lines for **(a)** precipitation, **(b)** streamflow of the three streams, **(c)** the groundwater in the riparian piezometers in each watershed, and **(d)** subsurface stormflow in the R watershed and groundwater. Note the different x and y axes in panel **(a)** vs. panels **(b–d)**.

−1.7 ‰ (^{18}O) and −26.7 and −14.7 ‰ (^{2}H) for the B stream, and −4.6 and −2.7 ‰ (^{18}O) and −24.4 and −17.2 ‰ (^{2}H) for the C stream (Fig. 3b–d, ^{2}H not shown). We attempted to fit an input–output transfer function between observed precipitation and runoff isotope ratios (McGuire and McDonnell, 2006) to determine catchment transit times. Only very poor fits were possible, suggesting that the transit time is much longer than the data series length and likely beyond the scope of naturally occurring stable isotopes, consistent with water balance calculations of stream transit time (Du et al., 2014).

3.2 Isotopic water lines of water cycle components

The δ^{2}H–δ^{18}O relation for precipitation, streamwater, groundwater, and subsurface stormflow is shown in Fig. 4 and summarized in Table 1. These data show that the slopes of each of these lines are systematically offset from local precipitation (Fig. 4a). The local meteoric water line (LMWL) and the global meteoric water line (GMWL) are compared in Fig. 5.

Throughfall (not shown) was slightly enriched compared to open precipitation. The slope of the throughfall water lines varied between 6.00 and 7.03 and intercepts between 5.51 and 8.96 for different locations. The EWLs of the of the three streams (Fig. 4b) showed very strong evaporative enrichment of heavy isotopes, based on measured slope and intercept as presented in Table 1. The EWLs of the riparian groundwater (Fig. 4c, Table 1) were very similar to the EWLs of the streams and showed the same strong enrichment. We do not present the regression relations between ^{18}O and ^{2}H for the samples of the two piezometers in the riparian zone of the B stream since the regression was not significant ($p > 0.05$).

Water collected as lateral subsurface stormflow (SSF) from the hillslope trench (shown in Fig. 1) in watershed R combined soil water and event precipitation. The EWLs of these mobile, shallow subsurface waters (Fig. 4d, Table 1) fell between the slope of precipitation and streamwater. Groundwater from the 14 wells also showed distinct evaporative enrichment (Fig. 4d, Table 1). We did not further differentiate the EWLs of different groundwater wells due to the low number of samples for each well. The δ^{18}O (and δ^{2}H, not shown) values of the riparian-zone water were closely linked to the values observed in the corresponding streams (Fig. 6). Especially in the R watershed, δ^{18}O from both piezometers was very similar to the observed values in the stream over the observation period (Fig. 6a). The same pattern was observed

Table 1. Summary of the MWLs/EWLs of the different hydrological compartments, including their regression equation, the coefficient of determination (R^2) and p value of the regression, and the number of samples available for the regression. R, B, and C stand for the three watersheds, stream indicates streamflow, riparian stands for the riparian groundwater, SSF for subsurface stormflow, and groundwater for the deeper groundwater at SRS.

Compartment	Regression equation	R^2	p value	Number of samples
LMWL	$\delta^2H = 7.15 \cdot \delta^{18}O + 9.28$ ‰	0.93	$\ll 0.01$	145
R stream	$\delta^2H = 2.52 \cdot \delta^{18}O - 11.88$ ‰	0.40	$\ll 0.01$	134
B stream	$\delta^2H = 2.86^* \cdot \delta^{18}O - 9.66$ ‰	0.78	$\ll 0.01$	38
C stream	$\delta^2H = 2.84 \cdot \delta^{18}O - 9.95$ ‰	0.55	$\ll 0.01$	76
R riparian	$\delta^2H = 2.09 \cdot \delta^{18}O - 14.89$ ‰	0.67	$\ll 0.01$	38
C riparian	$\delta^2H = 2.52 \cdot \delta^{18}O - 12.21$ ‰	0.62	$\ll 0.01$	35
SSF	$\delta^2H = 4.58 \cdot \delta^{18}O - 2.11$ ‰	0.75	$\ll 0.01$	22
Groundwater	$\delta^2H = 3.53 \cdot \delta^{18}O - 8.27$ ‰	0.45	$\ll 0.01$	117

Table 2. Values of p to evaluate the differences between the LMWL/EWLs of the different water compartments used to constrain the conceptual model.

	B stream	C stream	Riparian-zone C watershed	Groundwater	R stream	Precipitation	Riparian-zone R watershed	SSF
B stream	NA	0.58	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
C stream	0.58	NA	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Riparian-zone C watershed	< 0.01	< 0.01	NA	< 0.01	0.99	< 0.01	< 0.01	< 0.01
Groundwater	< 0.01	< 0.01	< 0.01	NA	< 0.01	< 0.01	< 0.01	< 0.01
R stream	< 0.01	< 0.01	0.99	< 0.01	NA	< 0.01	< 0.01	< 0.01
Precipitation	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	NA	< 0.01	< 0.01
Riparian-zone R watershed	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	NA	< 0.01
SSF	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	NA

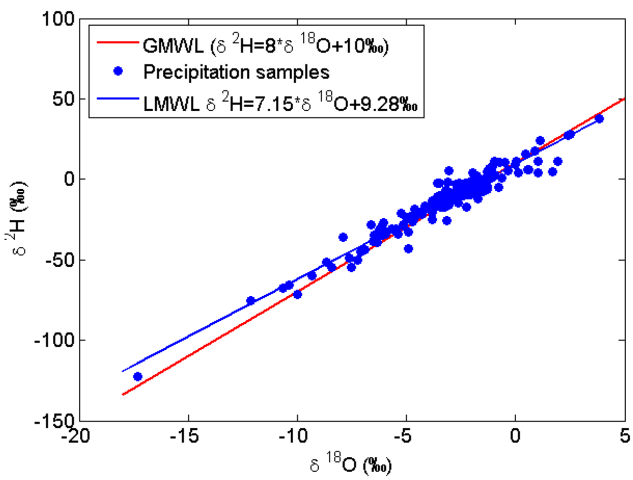

Figure 5. The Global Meteoric Water Line (GMWL) compared to the Local Meteoric Water Line (LMWL) of the study site.

in the B watershed (Fig. 6b), while the $\delta^{18}O$ values of riparian groundwater in the C watershed were often lower than the corresponding streamwater (Fig. 6c). During March 2011 some differences between the piezometers and the stream values were observable. Stream discharge was very low at this point so that some direct precipitation onto the channel itself may explain this effect. In March, we observed one precipitation sample with a very heavy $\delta^{18}O$ value of 3.9 ‰.

Further, it is important that the various compartments have significantly different EWLs. This would eventually allow to unambiguously differentiate between them. We used a two-sample t test to evaluate this. The results are summarized in Table 2 and indicate that most components are indeed significantly different from each other.

3.3 Isotopes of nitrate in water cycle components

The dual-isotope plot of $\delta^{18}O_{NO_3}$ vs. $\delta^{15}N_{NO_3}$ (Fig. 7) showed distinct differences in the signatures of nitrate in the different water cycle components in the R watershed. The signatures of the streamwater overlap with those of the riparian zone. In contrast, nitrate isotope signatures of subsurface stormflow from the trench can reach high values that approach the signatures in throughfall, suggesting a fast transformation of throughfall into subsurface stormflow.

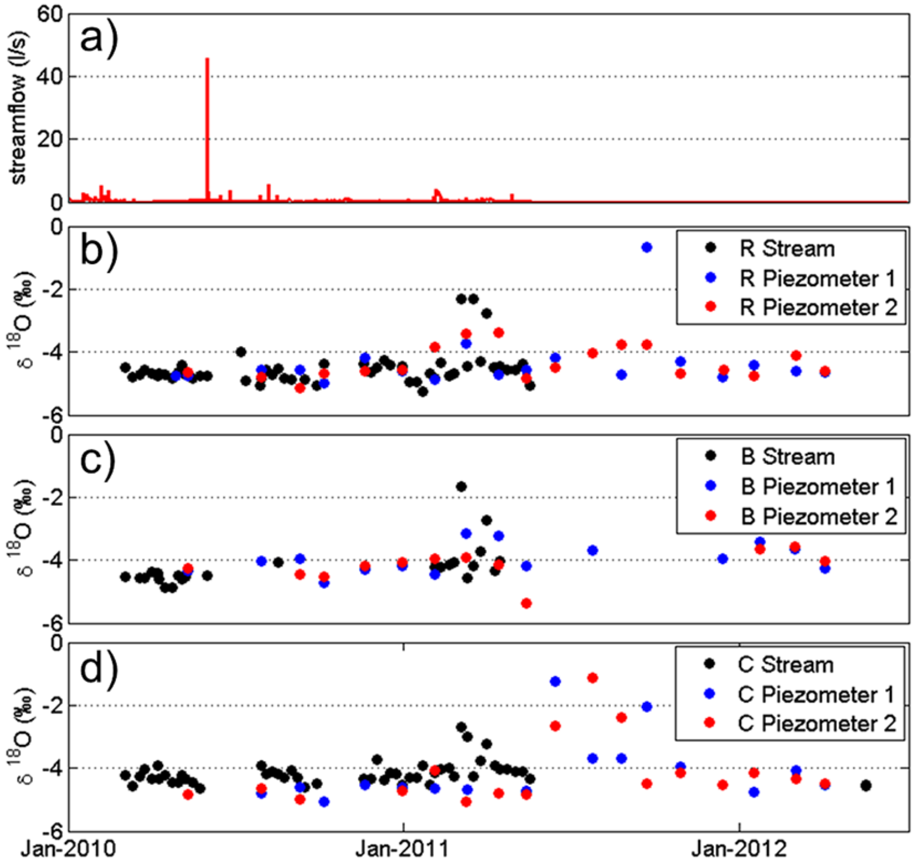

Figure 6. Temporal dynamics of the streamflow exemplified by the R stream **(a)** and of ^{18}O in riparian groundwater and the stream outlet for the R **(b)**, B **(c)**, and C **(d)** watersheds.

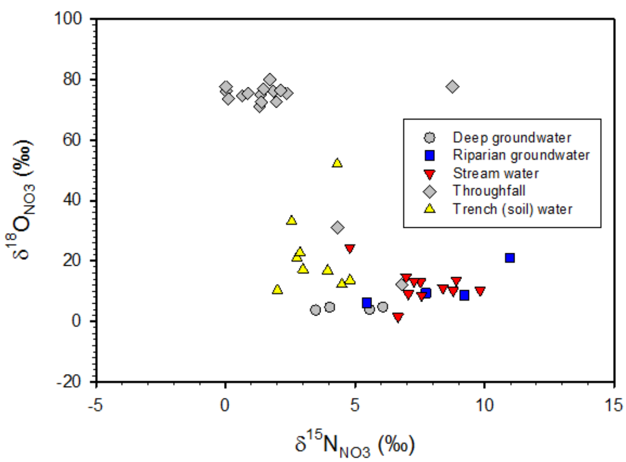

Figure 7. Biplot of $\delta^{15}N_{NO_3}$ and $\delta^{18}O_{NO_3}$ of nitrate (‰) in water samples collected from the intermittent stream (white square), riparian groundwater (black circle), throughfall (light grey triangle), deep groundwater (dark grey triangle), and trench flow water (grey diamond) in the R watershed.

4 Discussion

The three watersheds showed very low annual runoff ratios during the 3-year record, combined with long spells of zero flow. This is similar to the findings in Sun et al. (2002), who showed highly ephemeral stream discharge patterns for their coastal plain site. Like Amatya et al. (1996) and Slattery et al. (2006), we found that soil properties, especially buried argillic horizons with low permeability (i.e., the throttle for lateral flow), strongly influenced runoff generation in these low-relief coastal plain regions. In related work at our site, Du et al. (2014) observed that the trenched hillslope (draining 13 % of the R watershed) can generate higher discharge peaks than measured at the catchment outlet. For another catchment in the Atlantic Coastal Plain, La Torre Torres et al. (2011) showed the importance of evapotranspiration on runoff generation, due to its effect on water table position and its subsequent control on runoff. They also found a strong seasonality in runoff ratios based on the seasonality in evapotranspiration and rain amount during wet periods, consistent with the catchment behavior in our study.

Our site, like that reported by Devito et al. (2005a), showed that dry catchment conditions frequently led to dis-

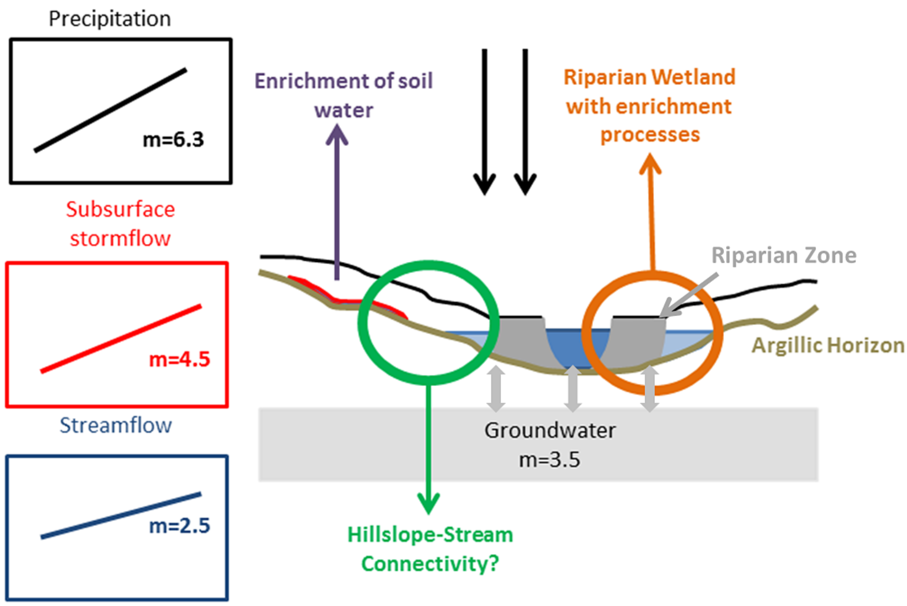

Figure 8. Conceptual model of baseflow runoff generation and enrichment in heavy isotopes from rainfall to streamflow. Key element is the disconnectivity between the hillslopes and the riparian-stream systems, which is likely sustained by precipitation and deeper groundwater.

connectivity of the uplands with the valley bottom and stream. This resulted in low runoff coefficients and the dominance of evaporation in the water balance. In addition, direct precipitation on the stream channel can alter the isotope signal, when flow is close to zero. This was observed during March 2011, when very heavy precipitation ($\delta^{18}O = 3.9$ ‰) led to a deviation between stream isotope signals and riparian isotopic signals adjacent to the streams throughout the area. Figure 8 conceptually summarizes the runoff generation and isotopic signature at the study site. A key element is the rare or nonexistent connectivity in the hillslope–riparian–stream continuum and the enrichment in heavy water isotopes in the riparian zone/wetlands that supplies baseflow. Further, the deeper groundwater system can interact with the groundwater of the riparian zone during wet conditions and is likely a major contributor to the riparian groundwater.

4.1 What do the slopes of different source components mean and are they realistic?

Evaporation between rain events had a significant effect on the isotopic composition of streamflow. Isotopic fractionation via evaporation leads to a stronger kinetic effect for ^{18}O compared to ^{2}H, resulting in evaporative enrichment of the water along an evaporation water line with a lower slope relative to the original water (Gonfiantini, 1986). While the variability in stream ^{18}O and ^{2}H is low over time, the isotope data exhibited a strong enrichment in heavy isotopes compared to precipitation and throughfall. Our samples of groundwater, subsurface stormflow, and streamflow all exhibited significant ($p \leq 0.05$) isotopic enrichment compared to the local precipitation. The observed slopes are lower than expected for South Carolina, based on the work of Gibson et al. (2008), who modeled a slope of 4–5 for open water bodies and 3–4 for soil water for the region. The strong evaporative enrichment of groundwater suggests groundwater recharge influenced by enriched soil water. Streams and riparian groundwater were even more enriched in heavy isotopes, suggesting further isotopic enrichment of the riparian groundwater as it reemerged in the low-relief and slow-moving stream floodplain. Our measured isotopic enrichment and the low annual runoff coefficients suggest that evapotranspiration strongly influences the runoff dynamics in the R, B, and C watersheds, consistent with the behavior of other lower-relief watersheds in the Atlantic Coastal Plain of the USA (La Torre Torres et al., 2011) and elsewhere (Devito et al., 2005a).

To our knowledge such shallow slopes for streamwater have not been reported in the literature. We think that measurement errors are unlikely since the slopes of the LMWL of our precipitation sample fit the expectations. The statistical significance of the relationship of ^{2}H and ^{18}O was significant ($p \leq 0.05$) for all three streams, indicating that these EWLs describe the streamflow. Furthermore, the removal of several relatively high isotopic values from the stream EWL (Fig. 4) does not significantly change the slope, suggesting the relationship is robust across the measured ^{18}O and ^{2}H values. Surface water sampled from two Carolina Bay wetlands also showed strong evaporative enrichment, suggesting that the observed stream EWLs are not simply a mixing line between an evaporative-groundwater and a rain-fed wetland that suddenly becomes connected to the stream outlet. Lower slopes than predicted by Gibson et al. (2008) could also derive from water vapor mixing processes between terrestrial and oceanic air masses leading to evaporation lines with lower slopes. Further work to explore the exceptionally low slopes is needed as this is an interesting phenomenon in and of itself.

4.2 The dual-isotope approach for conceptualizing flow sources in low-angled terrain

The use of stable isotopes of water has been a valuable tool for determining the geographic sources and temporal components of hydrographs (Klaus and McDonnell, 2013). When isotopes are combined with chemical tracers, they may also be useful for determining the importance of different landscape elements in the generation of flow at catchment scale (Burns et al., 2001; McGlynn and McDonnell, 2003; Ocampo et al., 2006). Key prerequisites for all of these approaches are distinct end-members and an isotope time series that deflects from pre-event conditions through time (Sklash and Farvolden, 1979). Our streamwater isotopic time series showed (with few exceptions) few deflections through time and, consequently, provided little insight into time- and source-components and hillslope–riparian–streamflow connectivity. Furthermore, our isotope time series did not yield a meaningful transit time estimate, suggesting that transit times are longer than the range used for stable isotopes, likely > 5 years.

In the low-relief watersheds at the SRS, where the classical methods of isotope hydrology are limited by the lack of temporal dynamics of the stable isotope time series, our dual-isotope approach was useful for determining the connectivity/disconnectivity between different water cycle components. The use of the individual water lines adds value to our understanding of runoff generation in this low-angled terrain and is consistent with hydrometric observations (Du et al., 2014) and nitrate stable isotopes. The use of the water line approach clarifies the close link between the groundwater, the riparian water, and the stream and shows that the riparian zone controls the isotopic composition of streamflow.

This method is useful to constrain the linkages in low-angled terrain but also allows additional insight into data-scarce catchments; this can provide a fundamental understanding of where water comes from. While the water line approach is able to constrain a general conceptual model (Fig. 8) of where water comes from, the approach exhibits clear limitations. The mixing of two water types with clearly different isotopic enrichments can lead to mixing lines in the resulting water that can interfere with a meaningful interpretation of the resulting water lines. The relative position of a sample along this mixing line indicates the contribution of multiple water sources with a different degree of evaporative enrichment. This will prohibit a quantitative mixing calculation based on the characteristics of the water lines for a distinct sample of streamwater. Nevertheless, the approach presented in this paper can clearly constrain where water comes from during baseflow conditions in a watershed at different antecedent conditions, confirmed by nitrate isotope data.

5 Conclusions

We examined the source of runoff in a set of lowland forested watersheds in South Carolina, USA. Streamflow was very ephemeral and the time series of the stable isotopic composition of streamwater showed minimal temporal dynamics compared to rainfall. Notwithstanding, our dual-isotope approach based on the water lines was able to isolate and separate hillslope, riparian and deep groundwater, and streamflow compositions. The streams in each of our watersheds showed strong evaporative enrichment compared to the local meteoric water line ($\delta^2\mathrm{H} = 7.15 \cdot \delta^{18}\mathrm{O} + 9.28$ ‰), with slopes of 2.52, 2.84, and 2.86. Based on the unique and unambiguous slopes of the EWLs of the different water cycle components and the isotopic time series of the individual components, we were able to show how the riparian zone controls baseflow in this system and how the riparian zone "resets" the stable-isotope composition of the observed streams in our low-angle, forested watersheds. Deeper groundwater likely supplies the riparian-groundwater system. These findings were supported by the overlap of nitrate stable isotope signatures ($^{18}\mathrm{O}_{\mathrm{NO_3}}$ and $^{15}\mathrm{N}_{\mathrm{NO_3}}$) of riparian groundwater and streamwater in the R watershed. Our approach allowed for a general description of long-term sources to streamflow, especially baseflow, even though in situ mixing calculations were not possible.

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References

Ali, G. A., L'Heureux, C., Roy, A. G., Turmel, M. C., and Courchesne, F.: Linking spatial patterns of perched groundwater storage and stormflow generation processes in a headwater forested catchment, Hydrol. Process., 25, 3843–3857, doi:10.1002/hyp.8238, 2011.

Amatya, D. M., Skaggs, R. W., and Gregory, J. D.: Effects of controlled drainage on the hydrology of drained pine plantations in the North Carolina coastal plain, J. Hydrol., 181, 211–232, doi:10.1016/0022-1694(95)02905-2, 1996.

Anderson, M. G. and Kneale, P. E.: The influence of low-angled topography on hillslope soil-water convergence and stream discharge, J. Hydrol., 57, 65–80, doi:10.1016/0022-1694(82)90103-2, 1982.

Bachmair, S. and Weiler, M.: New dimensions of Hillslope Hydrology, in: Forest Hydrology and Biogeochemistry, edited by: Levia, D. F., Carlyle-Moses, D., and Tanaka, T., Springer Netherlands, Amsterdam, 455–481, 2011.

Baer, D. S., Paul, J. B., Gupta, M., and O'Keefe, A.: Sensitive absorption measurements in the near-infrared region using off-axis integrated cavity output spectroscopy, International Symposium on Optical Science and Technology, Int. Soc. Opt. Photon., 167–176, 2002.

Branfireun, B. A. and Roulet, N. T.: The baseflow and storm flow hydrology of a precambrian shield headwater peatland, Hydrol. Process., 12, 57–72, 1998.

Burns, D. A., McDonnell, J. J., Hooper, R. P., Peters, N. E., Freer, J. E., Kendall, C., and Beven, K.: Quantifying contributions to storm runoff through end-member mixing analysis and hydrologic measurements at the Panola Mountain Research Watershed (Georgia, USA), Hydrol. Process., 15, 1903–1924, doi:10.1002/hyp.246, 2001.

Buttle, J. M., Dillon, P. J., and Eerkes, G. R.: Hydrologic coupling of slopes, riparian zones and streams: an example from the Canadian Shield, J. Hydrol., 287, 161–177, doi:10.1016/j.jhydrol.2003.09.022, 2004.

Casciotti, K. L., Sigman, D. M., Hastings, M. G., Böhlke, J. K., and Hilkert, A.: Measurement of the oxygen isotopic composition of nitrate in seawater and freshwater using the denitrifier method, Anal. Chem., 74, 4905–4912, doi:10.1021/ac020113w, 2002.

Devito, K. J., Hill, A. R., and Roulet, N.: Groundwater-surface water interactions in headwater forested wetlands of the Canadian Shield, J. Hydrol., 181, 127–147, doi:10.1016/0022-1694(95)02912-5, 1996.

Devito, K. J., Creed, I. F., and Fraser, C. J. D.: Controls on runoff from a partially harvested aspen-forested headwater catchment, Boreal Plain, Canada, Hydrol. Process., 19, 3–25, doi:10.1002/hyp.5776, 2005a.

Devito, K. J., Creed, I. F., Gan, T., Mendoza, C., Petrone, R., Silins, U., and Smerdon, B.: A framework for broad-scale classification of hydrologic response units on the Boreal Plain: is topography the last thing to consider?, Hydrol. Process., 19, 1705–1714, doi:10.1002/hyp.5881, 2005b.

Du, E., Jackson, C. R., McDonnell, J. J., Klaus, J., Griffiths, N. A., Williamson, M. F., Greco, J. L., and Bitew, M.: More fill, less spill: interflow behavior over a low-angle argillic layer, in review, 2014.

Eshleman, K. N., Pollard, J. S., and O'Brien, A. K.: Interactions between groundwater and surface water in a virginia coastal plain watershed, 1. Hydrological flowpaths, Hydrol. Process., 8, 389–410, doi:10.1002/hyp.3360080504, 1994.

Gibson, J. J., Birks, S. J., and Edwards, T. W. D.: Global prediction of δA and δ^2H–δ^{18}O evaporation slopes for lakes and soil water accounting for seasonality, Global Biogeochem. Cy., 22, GB2031, doi:10.1029/2007GB002997, 2008.

Gibson, J. J., Fekete, B. M., and Bowen, G. J.: Stable Isotopes in Large Scale Hydrological Applications, in: Isoscapes, Springer, Dordrecht, Heidelberg, London, New York, 389–405, 2010.

Gonfiantini, R.: Environmental isotopes in lake studies, Handbook of environmental isotope geochemistry, Elsevier, Amsterdam, 113–168, 1986.

Hitchcock, D. and Blake, J. I.: Effectiveness Monitoring Report MWMF Tritium Phytoremediation Interim Measures, Technical Report, USDA Forest Service, Savannah River, New Ellenton, SC, 2003.

Jencso, K. G., McGlynn, B. L., Gooseff, M. N., Wondzell, S. M., Bencala, K. E., and Marshall, L. A.: Hydrologic connectivity between landscapes and streams: Transferring reach- and plot-scale understanding to the catchment scale, Water Resour. Res., 45, W04428, doi:10.1029/2008WR007225, 2009.

Jencso, K. G. and McGlynn, B. L.: Hierarchical controls on runoff generation: Topographically driven hydrologic connectivity, geology, and vegetation, Water Resour. Res., 47, W11527, doi:10.1029/2011WR010666, 2011.

Kilgo, J. C. and Blake, J. I.: Ecology and management of a forested landscape: Fifty years on the Savannah River, Island Press, Washington, D.C., 2005.

Klaus, J. and McDonnell, J. J.: Hydrograph separation using stable isotopes: Review and evaluation, J. Hydrol., 505, 47–64, doi:10.1016/j.jhydrol.2013.09.006, 2013.

La Torre Torres, I. B., Amatya, D. M., Sun, G., and Callahan, T. J.: Seasonal rainfall-runoff relationships in a lowland forested watershed in the southeastern USA, Hydrol. Process., 25, 2032–2045, doi:10.1002/hyp.7955, 2011.

McGlynn, B. L. and McDonnell, J. J.: Quantifying the relative contributions of riparian and hillslope zones to catchment runoff, Water Resour. Res., 39, 1310, doi:10.1029/2003WR002091, 2003.

McGuire, K. J. and McDonnell, J. J.: A review and evaluation of catchment transit time modelling, J. Hydrol., 330, 543–563, doi:10.1016/j.jhydrol.2006.04.020, 2006.

Ocampo, C. J., Sivapalan, M., and Oldham, C.: Hydrological connectivity of upland-riparian zones in agricultural catchments: Implications for runoff generation and nitrate transport, J. Hydrol., 331, 643–658, doi:10.1016/j.jhydrol.2006.06.010, 2006.

Penna, D., Tromp-van Meerveld, H. J., Gobbi, A., Borga, M., and Dalla Fontana, G.: The influence of soil moisture on threshold runoff generation processes in an alpine headwater catchment, Hydrol. Earth Syst. Sci., 15, 689–702, doi:10.5194/hess-15-689-2011, 2011.

Rasmussen, T. C. and Mote, T. L.: Monitoring surface and subsurface water storage using confined aquifer water levels at the Savannah River Site, USA, Vadose Zone J., 6, 327–335, doi:10.2136/vzj2006.0049, 2007.

Rebel, K. T.: Using trees to remediate tritium contaminated groundwater: a modeling and tracer study, PhD thesis, Cornell University, Ithaca, NY, USA, 173 pp., 2004.

Redding, T. and Devito, K.: Mechanisms and pathways of lateral flow on aspen-forested, Luvisolic soils, Western Boreal Plains, Alberta, Canada, Hydrol. Process., 24, 2995–3010, doi:10.1002/hyp.7710, 2010.

Riha, S. and Rebel, K.: Savannah River Site Mixed Waste Management Facility Southwest Plume Tritium Phytoremediation Evaluating Irrigation Management Strategies Over 25 Years, Technical Report, USDA Forest Service, Savannah River, New Ellenton, SC, 2004.

Samuelson, L., Stokes, T., and Farris, M.: Measuring Transpiration to Regulate Winter Irrigation Rates, Final Report 06-27-R, USDA Forest Service, Savannah River, New Ellenton, SC, 2006.

Seibert, J. and McDonnell, J. J.: On the dialog between experimentalist and modeler in catchment hydrology: Use of soft data for multicriteria model calibration, Water Resour. Res., 38, 1241, doi:10.1029/2001WR000978, 2002.

Sidle, R. C., Tsuboyama, Y., Noguchi, S., Hosoda, I., Fujieda, M., and Shimizu, T.: Stormflow generation in steep forested headwaters: a linked hydrogeomorphic paradigm, Hydrol. Process., 14, 369–385, 2000.

Sigman, D. M., Casciotti, K. L., Andreani, M., Barford, C., Galanter, M., and Böhlke, J. K.: A bacterial method for the nitrogen isotopic analysis of nitrate in seawater and freshwater, Anal. Chem., 73, 4145–4153, doi:10.1021/ac010088e, 2001.

Sklash, M. G. and Farvolden, R. N.: The role of groundwater in storm runoff, J. Hydrol., 43, 45–65, doi:10.1016/0022-1694(79)90164-1, 1979.

Slattery, M. C., Gares, P. A., and Phillips, J. D.: Multiple modes of storm runoff generation in a North Carolina coastal plain watershed, Hydrol. Process., 20, 2953–2969, doi:10.1002/hyp.6144, 2006.

Sun, G., McNulty, S. G., Amatya, D. M., Skaggs, R. W., Swift Jr., L. W., Shepard, J. P., and Riekerk, H.: A comparison of the watershed hydrology of coastal forested wetlands and the mountainous uplands in the Southern US, J. Hydrol., 263, 92–104, doi:10.1016/S0022-1694(02)00064-1, 2002.

Todd, A. K., Buttle, J. M., and Taylor, C. H.: Hydrologic dynamics and linkages in a wetland-dominated basin, J. Hydrol., 319, 15–35, doi:10.1016/j.jhydrol.2005.05.001, 2006.

Wyatt, D. E. and Harris, M. K.: Overview of the history and geology of the Savannah River Site, Environ. Geosci., 11, 181–190, doi:10.1306/eg.06220404022, 2004.

Yi, Y., Gibson, J. J., Hélie, J. F., and Dick, T. A.: Synoptic and time-series stable isotope surveys of the Mackenzie River from Great Slave Lake to the Arctic Ocean, 2003 to 2006, J. Hydrol., 383, 223–232, doi:10.1016/j.jhydrol.2009.12.038, 2010.

Zehe, E., Elsenbeer, H., Lindenmaier, F., Schulz, K., and Blöschl, G.: Patterns of predictability in hydrological threshold systems, Water Resour. Res., 43, W07434, doi:10.1029/2006WR005589, 2007.

Zhang, Y., Shen, Y., Chen, Y., and Wang, Y.: Spatial characteristics of surface water and groundwater using water stable isotope in the Tarim River Basin, northwestern China, Ecohydrology, 6, 1031–1039, doi:10.1002/eco.1416, 2013.