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Exploring water cycle dynamics through sampling multitude stable water isotope pools in a small developed landscape of Germany

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Abstract

Conducting a dual stable water isotope (δ^2H and δ^{18}O) study in the developed landscape of the Schwingbach catchment (Germany) helped to unravel connectivity and disconnectivity between the different water cycle components. The two-year weekly to biweekly measurements of precipitation, stream, and groundwater isotopes revealed that surface and groundwater are decoupled from the annual precipitation cycle but showed bidirectional interactions between each other. Seasonal variations based on temperature effects were observed in the precipitation signal but neither reflected in stream nor in groundwater isotopic signatures. Apparently, snowmelt played a fundamental role for groundwater recharge explaining the observed differences to precipitation δ-values.

A spatially distributed snapshot sampling of soil water isotopes in two soil depths at 52 sampling points across different land uses (arable land, forest, and grassland) revealed that top soil isotopic signatures were similar to the precipitation input signal. Preferential water flow paths occurred under forested soils explaining the isotopic similarities between top and subsoil isotopic signatures. Due to human-impacted agricultural land use (tilling and compression) of arable and grassland soils, water delivery to the deeper soil layers was reduced, resulting in significant different isotopic signatures. However, the land use influence smoothed out with depth and soil water approached groundwater δ-values. Seasonally tracing stable water isotopes through soil profiles showed that the influence of new percolating soil water decreased with depth as no remarkable seasonality in soil isotopic signatures was obvious at depth >0.9 m and constant values were observed through space and time.

Little variation in individual isotope time series of stream and groundwater restricted the use of classical isotope hydrology techniques e.g. mean transit time estimation or hydrograph separation. Still, tracing stable water isotopes through the water cycle was valuable for determining interactions between different water cycle components and

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gaining catchment specific process understanding in a developed, human-impacted landscape.

1 Introduction

The application of stable water isotopes as natural tracers in combination with hydrodynamic methods has been proven to be a valuable tool for studying the origin, formation, and interrelationship between surface water and groundwater (Blasch and Bryson, 2007; Goni, 2006), partitioning evaporation and transpiration (Phillips and Gregg, 2003; Rothfuss et al., 2010, 2012; Wang and Yakir, 2000), and further mixing processes between various water sources (Aggarwal et al., 2007; Clark and Fritz, 1997; Kendall and Coplen, 2001; Wu et al., 2012). Particularly in catchment hydrology, stable water isotopes play a major role since they can be utilised for hydrograph separations (Buttle, 2006; Hoeg et al., 2000; Ladouche et al., 2001; Munyaneza et al., 2012), to calculate the mean transit time (Garvelmann et al., 2012; McGuire et al., 2002, 2005; Rodgers et al., 2005b), to investigate water flow paths (Barthold et al., 2011; Goller et al., 2005; Rodgers et al., 2005a), or to improve hydrological model simulations (Birkel et al., 2010; Koivusalo et al., 1999; Liebminger et al., 2007; Rodgers et al., 2005b). However, spatio-temporal sources of stream water in low angle, developed catchments are still poorly understood. This is partly caused by damped stream water isotopic signatures excluding traditional hydrograph separations (Klaus et al., 2014). Unlike the distinct watershed components found in steeper headwater counterparts, lowland areas often exhibit a complex groundwater–surface water interaction (Klaus et al., 2014). Moreover, the complex character of developed, agricultural dominated catchments is often disregarded and established research approaches often failed to fully capture agro-ecosystem functioning at multiple scales (Orlowski et al., 2014). This is mainly because almost all European river systems were already substantially modified by humans before river ecology research developed (Klapper, 1990; Allan, 2004). While agricultural land use (arable land, permanent crops, and grassland) is the most dominant land use in Europe (UNEP, 2002), few hydrological and especially stable water isotope studies have been conducted in these human-impacted landscapes and catchments. The challenges of future research are therefore to gain knowledge about the water pathways and interplay of the water cycle components in developed landscapes to improve our understanding of biogeochemical fluxes such as nitrogen compounds or pesticides in these managed landscapes (Orlowski et al., 2014).

To explore the relationship between precipitation, stream, soil, and groundwater detailed knowledge of the isotopic composition of the various water sources and their variation in space and time is required. In principal, isotopic signatures of precipitation are altered by temperature, amount (or rainout), continental, altitudinal, and seasonal effects. They are mainly influenced by prevailing atmospheric conditions during rainfall and snowfall causing a depletion of isotopes (Araguas-Araguas et al., 2000; Blasch and Bryson, 2007; Clark and Fritz, 1997; Gat, 1996; Kendall and McDonnell, 1998). The input signal becomes more pronounced in snow-dominated systems where snowfall and snowmelt are depleted in heavy stable water isotopes relative to rainfall (Maule et al., 1994; O'Driscoll et al., 2005). Stream water isotopic signatures can reflect precipitation isotopic composition and moreover, depend on discharge variations affected by seasonally variable contributions of different water sources such as bidirectional water exchange with the groundwater body during baseflow, or high event-water contributions during stormflow (Kendall and McDonnell, 1998; Koeniger et al., 2009). To distinguish between the direct contribution of precipitation input and groundwater to storm runoff, isotope hydrograph separation can be applied (Buttle, 1994, 2006; Cey et al., 1998). Furthermore, if the isotopic composition of the input signal is known, understanding of recharge areas (altitude), times of recharge (season) as well as residence times and influencing processes (e.g. evaporation and mixing) can be gained (Koeniger et al., 2009).

Following the way of precipitation over the unsaturated zone to the groundwater, the process of infiltration in itself is known to be a non-fractionating process (Kendall and McDonnell, 1998), except for mixing between different water pools (e.g. moving and

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standing water) (Gat, 1996). However, precipitation falling on vegetated areas is intercepted by plants and re-evaporated isotopically fractionated. The remaining throughfall – less in volume in comparison to the total rain input – infiltrates slower and can be affected by evaporation resulting in an enrichment of heavy isotopes, particularly in the upper soil layers (Kendall and McDonnell, 1998). The amount of water lost by evaporation, transpiration, and interception and thereby the magnitude of isotopic change mainly depends on climate and vegetation cover (Kendall and McDonnell, 1998). In the soil, specific isotopic profiles develop, characterized by an evaporative layer near the surface especially under arid and semi-arid climate. This decreases exponentially with depth (Zimmermann et al., 1968), representing a balance between the upward convective flux and the downward diffusion of the evaporative signature (Barnes and Allison, 1988). This early observation (Zimmermann et al., 1968) has been broadly used to determine the infiltration profile and rate of evaporation in arid and semi-arid regions (Brunner et al., 2008). However, in humid and semi-humid areas, this exponential decrease is generally interrupted by the precipitation isotopic signal. Hence, the combination of the evaporation effect and the precipitation isotopic signature determine the isotope profile in the soil (Song et al., 2011). Once soil water reaches the saturated zone, this isotope information is finally transferred to the groundwater (Song et al., 2011). Soil water can therefore be seen as a link between precipitation and groundwater, and the dynamics of isotopic composition in soil water are indicative of the processes of precipitation infiltration, evaporation of soil water, and recharge to groundwater (Blasch and Bryson, 2007; Song et al., 2011). They can further be understood as a long term average of rain events (Clark and Fritz, 1997).

To compare different water sources on the catchment-scale, local meteoric water lines (LMWL) are used. They represent the linear relationship between δ^2H and δ^{18}O of meteoric waters (Kendall and McDonnell, 1998) in contrast to the global meteoric water line (GMWL), which describes the world-wide average stable isotopic composition in precipitation (Craig, 1961a). Thus, the comparison of stable isotope data from stream, soil, or groundwater samples relative to the global or local meteoric water lines

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can provide general understandings on water cycle processes at specific research sites (Song et al., 2011).

Identifying the origin of water vapour sources and moisture recycling (Gat et al., 2001; Lai and Ehleringer, 2011), the deuterium-excess (d-excess), defined by Dansgaard (1964) as $d = \delta^2\text{H} - 8 \times \delta^{18}\text{O}$ can be used, since the d-excess mainly depends on the mean relative humidity of the air masses formed above the ocean surface (Zhang et al., 2013). In addition, the d-excess reflects the prevailing conditions during evolution, interaction, or mixing of air masses en route to the precipitation site (Froehlich et al., 2002). Consequently, the d-excess has been used as a powerful tool to reconstruct temporal changes in moisture supply for a given location (Feng et al., 2009).

In order to improve our understanding of developed landscapes, we performed a multi-method hydrodynamic-based study in the observation area of the Schwingbach catchment (Germany) first. Results obtained through this former study imply that the catchment is highly responsive indicated by fast runoff responses to precipitation inputs (Orlowski et al., 2014). Moreover, groundwater reacted with raising head levels to precipitation events almost as quickly as stream water. We further showed that streamflow was generated in the catchment headwater area and that gaining and losing stream reaches occurred in parallel along the studied stream affected by the underlying geology. To back up the results obtained by Orlowski et al. (2014) on the level of hydrological processes, we investigated stable water isotope pools and fluxes in the same catchment. Stable water isotopes in combination with hydrodynamic data of a two-year monitoring period (July 2011–July 2013) were utilised to test the following hypotheses:

1. Highly responsive rainfall–runoff behaviour results in strong temporal variation of stream water isotopic signatures.

2. The rainfall isotopic signature is quickly transferred to the groundwater, since fast groundwater head level rises are attributed to rainfall events.

3. Due to different flow paths of groundwater along the stream, distinguished groundwater isotopic signatures can be found.

Exploring the spatio-temporal isotopic variations in precipitation, stream, soil, and groundwater will further help to establish a local meteoric water line and investigate the transformations from precipitation to soil and groundwater. Moreover, the effect of small-scale landscape characteristics such as topographic wetness index (TWI), distance to stream, and vegetation cover on soil water isotopic composition – as interface between precipitation and groundwater – will be examined for testing hypothesis (2).

2 Materials and methods

2.1 Study area

The research was carried out in the Schwingbach catchment (Germany). The Schwingbach and its tributary the Vollnkirchener Bach are low-mountainous creeks (Fig. 1) with an altered physical structure of the stream system (channelled stream reaches, pipes, drainage systems, fishponds). Almost 46 % of the catchment is forested, which slightly exceeds agricultural land use (35 %) (Fig. 1a). Grassland (10 %) is mainly distributed along streams and smaller meadow orchards are located around the villages. The catchment encompasses an area of 9.6 km^2, with an altitude range from 233–415 m a.s.l. The Vollnkirchener Bach tributary is about 4.7 km in length and drains a 3.7 km^2 subcatchment area, which ranges in elevation from 235–351 m a.s.l. (Fig. 1a).

The Schwingbach catchment is underlain by argillaceous shale in the northern parts, serving as aquicludes (Mazor, 2003). Graywacke zones with lydit in the central, as well as limestone, quartzite, and sandstone regions in the headwater area provide aquifers with large storage capacities (Marinos et al., 1997; Mazor, 2003) (Fig. 1d). Loess covers Paleozoic bedrock at north- and east bounded hillsides (Fig. 1c). Streambeds consists of sand and debris covered by loam and some larger rocks (Lauer et al., 2013). Many downstream sections of both creeks are framed by armor stones (Orlowski et al., 2014).

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Figure 1c shows that the dominant soil types in the study area are Stagnosols (41 %) and mostly forested Cambisols (38 %). Stagnic Luvisols with thick loess layers are under agricultural use. The same is true for Regosol, Luvisols, and Anthrosols, which encompass an area of 7 %. Gleysols are found predominantly under grassland sites along the creeks.

The climate is classified as temperate with a mean annual temperature of 8.2 °C. An annual precipitation sum of 633 mm (for the hydrological year 1 November 2012–31 October 2013) was measured at the climate station at the catchment outlet (site 13, Fig. 1b). Discharge peaks from December to April (> 114 L s^{-1}) and low flows occur from July until November. Significant snowmelt peaks were observed during December 2012 and February 2013. Furthermore, May 2013 was an exceptional wet month. Likewise, characterised by discharge > 114 L s^{-1}. A more detailed description of runoff characteristics, especially for the Vollnkirchener Bach is given in Orlowski et al. (2014).

2.2 Monitoring network and water isotope sampling

The monitoring network consists of an automated weather station at the catchment outlet (Campbell Scientific Inc., AQ5, UK; equipped with a CR1000 data logger collecting air temperature at 2 m height, wind speed and direction, relative humidity, and solar radiation), three tipping buckets, 15 precipitation collectors, six stream water sampling points, and 22 piezometers (Fig. 1a and b). Precipitation data were corrected according to Xia (2006).

Two stream water sampling points (sites 13 and 18) in the Vollnkirchener Bach are installed with trapezium shaped RBC-flumes for gauging discharge (Eijkelkamp Agrisearch Equipment, Giesbeek, NL), and a V-weir is located at sampling point 64. RBC-flumes and V-weir are equipped with Mini-Divers® (Eigenbrodt Inc. & Co. KG, Königsmoor, DE) for automatically recording water levels and deriving continuous discharge data through the given stage-discharge relationships (Eijkelkamp, 2013). Discharge at the remaining stream sampling points was manually measured applying the salt dilution method (WTW-cond340i, WTW, Weilheim, DE), which can be precise to

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±5 % (Day, 1976; Moore, 2004). The 22 piezometers situated between the conjunction of the Schwingbach with the Vollnkirchener Bach and the upper RBC-flume of the Vollnkirchener Bach (site 18) are made from perforated PVC tubes sealed with a bentonite clay at the upper part of the tube to prevent contamination by surface water. For monitoring shallow groundwater levels, either combined water level/temperature loggers (Odyssey Data Flow System, Christchurch, NZ) or Mini-Diver® water level loggers (Eigenbrodt Inc. & Co. KG, Königsmoor, DE) are installed. Accuracy of Mini-Diver® is ±5 mm and for Odyssey data logger ±1 mm. For calibration purposes, groundwater levels are additionally measured manually via an electric contact gauge.

Stable water isotope samples of rainfall, stream-, and groundwater were taken over a two-year observation period (July 2011–July 2013) approximately on weekly intervals, except for the winter period. Snow samples were taken in winter 2012–2013. Each precipitation collector was made from a 1 L glass bottle prepared with a circular funnel of 0.10 m in diameter. Funnels were covered with a mosquito net to keep out leaves, insects, or windblown debris. Bottles were placed in PVC tubes to avoid heating, screwed to wooden pales, and installed 1 m above ground. To avoid sample evaporation, a table tennis ball was placed into each funnel and two layers of small plastic balls were inserted into the glass bottles (Windhorst et al., 2013).

Stream water samples were taken as grab samples at six locations – three sampling points at each stream (Vollnkirchener Bach sites: 13, 18, and 94; Schwingbach sites: 11, 19, and 64) (Fig. 1a and b). To account for possible spatial variation in groundwater, grab samples were collected from 17 piezometers (Fig. 1b). Since spatial variations between the piezometers under meadow was small, the amount of sampled piezometers was reduced to three sampling points under meadow (sites 1, 6, and 21), five under the arable field (sites 25–29), and four beside the Vollnkirchener Bach (sites 24, 31, 32, and 35). Additionally, a drainage pipe (site 15) located ~ 226 m downstream of site 18 was sampled. According to IAEA standard procedures, all samples were filled and stored in 2 mL brown glass vials covered by silicone septa (Mook, 2001).

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2.3 Isotopic soil sampling

2.3.1 Spatial variability

In order to analyse the effect of small-scale characteristics such as distance to stream, TWI, and land use on soil isotopic signatures as connecting compartment between precipitation and groundwater, we sampled a snapshot of 52 points evenly distributed over a 200 m grid around the Vollnkirchener Bach. Soil samples were taken at four consecutive rainless days (1–4 November 2011). The TWI was chosen as it combines topography and slope, which were assumed to have an impact on soil water isotopic signatures (Garvelmann et al., 2012). Distances to the stream are linked to water flow path lengths and were therefore supposed to be a controlling factor. Vegetation has already been proven to have an impact on soil water isotopes (Brodersen et al., 2000; Gat, 1996; Li et al., 2007), and hence was expected to be a controlling factor as well.

Sampling sites were selected via a stratified, GIS-based sampling plan (ArcGIS, Arc Map 10.2.1, Esri, California, USA), including three classes of topographic wetness indices (TWIs: 4.4–6.5; 6.5–7.7; 7.7–18.4), two different distances to stream (0–121, 121–250 m), and three land use units (arable land, forest, and grassland), with each class containing the same number of sampling points. Samples were collected at depths of 0.2 and 0.5 m. Gravimetric water content was measured according to DIN-ISO 11465 by drying soils for 24 h at 110 °C. Soil pH was analysed following DIN-ISO 10390 on 1 : 1 soil-water-mixture with a handheld pH-meter (WTW cond340i, WTW Inc., DE). Bulk density was determined according to DIN-ISO 11272, and soil texture by finger testing (Brajendra et al., 2007; Whitefield, 2004).

2.3.2 Seasonal isotope soil profiling

In order to trace the seasonal development of stable water isotopes from rainfall to groundwater, seven soil profiles were taken in the dry summer season (28 August 2011), seven in the wet winter period (28 March 2013), and two profiles in the transitional season spring (14 April 2013) under different vegetation cover (arable land and grassland). Soil was sampled utilising a hand-auger (Eijkelkamp Agrisearch Equipment BV, Giesbeek, DE). Samples were taken from the soil surface to 2 m. Samples were collected in greater detail near the soil surface since this area is known to have the greatest isotopic variability (Barnes and Allison, 1988; Hsieh et al., 1998; Zimmermann et al., 1968).

Soil samples were stored in amber glass tubes, sealed with Parafilm®, and kept frozen until water extraction (Orlowski et al., 2013). Soil water was extracted cryogenically with 180 min extraction duration, a vacuum threshold of 0.3 Pa, and an extraction temperature of 90 °C. Isotopic signatures of δ^{18}O and δ^{2}H were analysed following the IAEA standard procedure (Newman et al., 2009) via off-axis integrated cavity output spectroscopy (OA-ICOS) (DLT-100, Los Gatos Research Inc., Mountain View, CA, USA). Isotopic ratios are reported in per mil (‰) relative to Vienna Standard Mean Ocean Water (VSMOW) (Craig, 1961b):

$$\delta^{2}\mathrm{H} \quad \text{or} \quad \delta^{18}\mathrm{O} = \left(\frac{R_{\text{sample}}}{R_{\text{standard}}} - 1\right) \times 1000 \qquad (1)$$

Here, R_{sample} and R_{standard} are ^{2}H/^{1}H or ^{18}O/^{16}O ratios of the sample and standard, respectively. Accuracy of analyses was 0.6 ‰ for δ^{2}H and 0.2 ‰ for δ^{18}O (LGR, 2013).

2.4 Statistical analyses

For statistical analyses, we used IBM SPSS Statistics (Version 22, SPSS Inc., Chicago, IL, US). Studying temporal and spatial variations in meteoric and groundwater, isotope data were tested for normal distribution. Subsequently, t tests or Multivariate Analyses of Variances (MANOVAs) were applied and Tukey-HSD tests were run to determine which groups were significantly different ($p \leq 0.05$). Event mean values of isotopes in precipitation, stream, and groundwater were calculated when no spatial variation was observed.

For comparisons, precipitation isotope data from the closest GNIP (Global Network of Isotopes in Precipitation) station Koblenz (DE; 73.8 km SW of the study area, 97 m a.s.l.) was used (IAEA, 2014). For monthly mean calculations, e.g. monthly d-excess values of the GNIP station Koblenz and its comparison to the Schwingbach event mean d-excess; the utilised data set comprises 24 values starting from July 2003–July 2005. For the calculation of the LMWL of Koblenz, the complete available GNIP data set was used (monthly means from 1 March 1981–1 December 2005; $N = 294$ values).

3 Results and discussion

Descriptive statistics of isotopic composition in precipitation, stream-, and groundwater are shown in Table 1 and are described in detail in the following:

3.1 Isotopes of precipitation

The δ^2H values of all precipitation isotope samples ($N = 592$) taken throughout the observation period (July 2011–July 2013) ranged from −167.6 to −8.3 ‰. To examine the spatial isotopic variation in precipitation, open rainfall was collected at 15 locations throughout the Schwingbach main catchment (Fig. 1a and b) for a 7 month period. Mook et al. (1974) observed for north-western Europe that the δ^{18}O values of precipitation collected over periods of 8 and 24 h from three locations within 6 km^2 at the same altitude were consistent within 0.3 ‰ (Gat et al., 2001). Likewise, no spatial variation could be observed in the Schwingbach catchment. Thus, rainfall was collected at the catchment outlet (site 13) from 23 October 2014 onward and event mean δ-values were calculated for the previous isotope data.

Analysing effects that influence the isotopic composition of precipitation, neither an amount effect nor an altitude effect was found – not surprisingly, as the greatest altitudinal difference between sampling points was only 101 m. Nevertheless, a slight tem-

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perature effect ($R^2 = 0.54$ for δ^2H and $R^2 = 0.61$ for δ^{18}O, respectively) was observed showing enriched isotopic signatures at higher temperatures (Fig. 2).

The linear relationship between air temperature (T in °C) and δ^{18}O values in precipitation at the Schwingbach catchment $\delta^{18}\text{O} = 0.44\,T - 12.05\,‰$ compares reasonably well with a correlation reported by Yurtsever (1975) based on north Atlantic and European stations from the GNIP network $\delta^{18}\text{O} = (0.521 \pm 0.014)\,T - (14.96 \pm 0.21)\,‰$. Rozanski et al. (1982) calculated $\delta^2\text{H} = (2.4 \pm 0.3)\,T - (80.5 \pm 4.2)\,‰$ ($R^2 = 0.89$) at the GNIP station Stuttgart, which is located 196 km South of the Schwingbach study area. This relationship is likewise similar to the correlation found for the Schwingbach catchment (Fig. 2). However, 53 % of the events were sampled at daily mean temperatures > 10 °C, resulting in a slight overrepresentation of values measured at warmer temperatures. Nevertheless, such a correspondence between the degree of isotope depletion and the temperature reflects the influence of the temperature effect in the catchment, which mainly appears in continental, middle–high latitudes (Jouzel et al., 1997; Wu et al., 2012). Furthermore, the correlation between δ^2H in monthly precipitations and local surface air temperature becomes increasingly stronger towards the centre of the continent (Rozanski et al., 1982).

Strong temporal variations in precipitation isotopic signatures, as well as pronounced seasonal isotopic effects were measured with greatest isotopic differences occurring between summer and winter. Samples taken in the fall and spring were isotopically similar, however differed from winter isotopic signature, which were somewhat lighter (Fig. 3). Furthermore, in the winter of 2012–2013 snow could be sampled, which decreased the mean winter isotopic values for this period in comparison to the previous winter period (2011–2012). No statistically significant inter-annual variation was detected between the summer periods of 2011 and 2012 (Fig. 3), which could have reflected varying local climate conditions (Koeniger et al., 2009). Inter-seasonal differences were mainly attributed to seasonal differences in air temperature and water vapour and their effect on evaporation (Schürch et al., 2003) and the presence of snow in the winter of 2012–2013 (Fig. 3). This observation is well in agreement with Gat et al. (2001) who stated that for temperate climates the $\delta^{18}O$ values generally do not vary by more than 1 ‰ inter-annually, and a large part of the spread is caused by variations in the average annual temperature. Moreover, the interior of the continent is obviously far more stable with regard to isotopic inputs than areas under greater influence of the Atlantic. Perhaps in view of this stability, only few isotope data are available for this region, apart from the general GNIP-maps (Bowen and Wilkinson, 2002; Darling, 2004; IAEA, 2014), for which this work contributes some valuable information.

3.1.1 Deuterium-excess

Examining the influence of moisture recycling on the isotopic compositions of precipitation, the deuterium-excess (d-excess) was calculated for each individual rain event at the Schwingbach catchment. For the two-year observation period, d-excess values ($N = 108$) ranged from −7.8 to +19.4 ‰ and averaged +7.1 ‰ (Fig. 3). In general, 37 % of all events were sampled in summer periods (21 June–21/22 September) and showed lower d-excess values in comparison to the 19 % winter precipitation events (21/22 December–19/20 March) (Fig. 3). D-excesses greater than +10 ‰ were determined for 22 % of all events. It is well-known that d-excess values for precipitation events originating from oceanic moisture are close to +10 ‰ (Craig, 1961a; Dansgaard, 1964; Wu et al., 2012). As a reference the d-excess of the GMWL $d = 10$ is depicted in Fig. 3 (dashed line).

Lowest values corresponded to summer precipitation events with evaporation of the raindrops below the cloud base at mean daily air temperatures between 12–18 °C. Same observations were made by Rozanski et al. (1982) for European GNIP stations. Accordingly, even more negative summer d-excess values were measured at air temperatures around 26–27 °C for a study site in Greece (Argiriou and Lykoudis, 2006).

Most of the higher values (> +10 ‰) appeared in cold seasons (fall/winter) (Fig. 3) similar to d-excess values observed by Wu et al. (2012) for a continental, semi-arid study area in Inner Mongolia (China). Winter snow samples of the Schwingbach catchment with very depleted δ-values showed highest d-excess values, which was again well in agreement with results of Rozanski et al. (1982) for European GNIP stations.

In comparison with the GNIP station Koblenz, the mean annual d-excess at the Schwingbach catchment was almost 5 ‰ higher (7.1 ‰ for 2011–2012 and 2012–2013, respectively), showing a greater impact of oceanic moisture sources than station Koblenz. Continental precipitation events originating from oceanic moisture can approach d-excess values of +10 ‰ (Wu et al., 2012) (Fig. 3, dashed line). Mean annual d-excess at the GNIP station Koblenz was 3.1 ‰ for 2003–2004 and 2.4 ‰ for 2004–2005. Nevertheless, highest d-excesses at the GNIP station matched highest values in the Schwingbach catchment, both occurring in the cold season. Differences in d-excess values between the Schwingbach catchment and the GNIP station Koblenz can be attributed to the fact that different observation periods were considered and likewise different climatic settings.

An amount effect on the d-excess has most likely been detected in the tropics (Bony et al., 2008) or for intense convective rain events (Gat et al., 2001) at monsoon-dominated sites (Risi et al., 2008). Since no amount effect on the δ^2H and δ^{18}O values was observed in the Schwingbach, also no linear regression of event d-excess with precipitation amount was detected.

3.2 Isotopes of stream water

In order to prove hypothesis (1) stream water isotope data were examined for their spatio-temporal variation. Analysing spatial differences in isotopic compositions between Schwingbach (sites 11, 19, and 64) and Vollnkirchener Bach (sites 13, 18, and 94) stream water resulted in no statistically significant differences for all sampling points (Fig. 4). Examining temporal isotopic variations, damped seasonality (less variation) of the isotope concentration in stream water in comparison to precipitation was measured with main seasonal differences occurring between summer and winter periods (Fig. 4). There was no evidence of rainfall affected stream water isotopic signatures throughout the two-year study period (Fig. 4) leading to the rejection of hypothesis (1). However,

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outlying depleted stream water isotopic signatures (e.g. in March 2012 and 2013) especially at site 64 can be explained by snowmelt (Fig. 4). In general, $\delta^{18}O$ values varied for the Vollnkirchener Bach by -8.4 ± 0.4 ‰ and for the Schwingbach by -8.4 ± 0.6 ‰ over the two-year observation period (Table 1). Schürch et al. (2003) likewise observed damped river water isotopic signatures as compared with precipitation isotopic signatures for sampling points of the "Swiss National Network for the Observation of Isotopes in the Water Cycle". For larger rivers like the Elbe at Torgau in eastern Germany seasonal isotopic composition varied with an amplitude of 1.5 ‰ in $\delta^{18}O$ (Darling, 2004).

Stream water isotopic signatures in the Schwingbach catchment were by approximately -15 ‰ in $\delta^{2}H$ more depleted than precipitation signatures (Table 1). However, surface water isotopic compositions were similar to groundwaters (Table 1), assuming that groundwater predominantly feeds baseflow. Even during peak flow occurring in January 2012, December–April or May 2013, rainfall input does not play a major role for stream water isotopic composition although fast rainfall–runoff behaviours were observed by Orlowski et al. (2014).

In conclusion, stream water isotopic time series of the Vollnkirchener Bach and Schwingbach showed (with few exceptions) little deflections through time and, consequently, provided little insight into time and source-components connectivity. Klaus et al. (2014) likewise had difficulties to apply traditional methods of isotope hydrology (mean transit time estimation, hydrograph separation) to their dataset due to the lack of temporal isotopic variation in stream water of a forested low-mountainous catchment in South Carolina (USA). However, little information is available for developed low-angle catchments so far. Furthermore, our and their isotope time series did not yield a meaningful transit time estimation (results not shown), suggesting that transit times are longer than the range used for stable water isotopes, likely > 5 years (Klaus et al., 2014).

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3.3 Isotopes of groundwater

Since groundwater head levels responded almost as quickly as streamflow to rainfall events, rainfall isotopic signatures were assumed to be rapidly transferred to the groundwater. This hypothesis (2) was likewise underlined by the fact that Orlowski et al. (2014) observed bidirectional water interactions between the groundwater body and the stream. Studying groundwater isotopic signatures at the downstream section of the Vollnkirchener Bach, almost constant isotopic values (Fig. 5, Table 1) throughout the study period were observed (δ^2H: -57.58 ± 1.6 ‰ for piezometers under meadow). Most depleted groundwater isotopic values (< -80 ‰ for δ^2H) were measured for piezometer 32 during snowmelt events in March and April 2013. Meltwater is known to be depleted in stable isotopes as compared to the annual mean of precipitation or groundwater (Kendall and McDonnell, 1998). In the Schwingbach catchment, groundwater under meadow differed from mean precipitation values by about -14 ‰ for δ^2H showing no evidence of a rapid transfer of rainfall isotopic signatures to the groundwater. As groundwater isotopic values are less variable through time, they rather seemed to be a mixture of former lighter precipitation events and snowmelt. We therefore assume that groundwater is mainly recharged throughout the winter. Likewise, O'Driscoll et al. (2005) showed that summer precipitation does not significantly contribute to recharge in the Spring Creek watershed of central Pennsylvania (USA) since δ^{18}O values in summer precipitation were enriched compared to mean annual groundwater composition. The hypothesis (2) of a quick transfer of recent rainfall isotopic signatures to the groundwater could therefore be falsified.

Due to different water flow paths of groundwater along the studied stream, distinguished groundwater isotopic signatures were hypothesised (3) to be found.

In fact, we could identify spatial statistical differences between grassland and arable land groundwater isotopic signatures. Groundwater isotopic signatures under arable land (sites: 25–29, Fig. 1b) showed more enriched values (Fig. 6). Isotopic signatures within piezometers under arable land varied among themselves, indicating hydrologi-

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cal disconnectivity between each other and the Vollnkirchener Bach as already stated by Orlowski et al. (2014). In contrast, the δ^2H and δ^{18}O values of the piezometers located beside the Vollnkirchener Bach (sites: 24, 31, 32, and 35) were statistically similar to the mean groundwater isotopic composition measured under meadow (sites: 3, 6, and 21). Moreover, groundwater isotopes under meadow were in close relation to stream water isotopes (Table 1). Orlowski et al. (2014) showed that influent and effluent conditions occurred simultaneously at different stream sections of the Vollnkirchener Bach affecting stream and groundwater isotopic compositions, likewise. Since groundwater head levels in the Vollnkirchener Bach subcatchment closely followed stream runoff-dynamics and responded to stormflow events with rising head levels (Fig. 5), we conclude that bidirectional water exchange between the groundwater body and the Vollnkirchener Bach occurred. However, both water compartments differed significantly from rainfall isotopic signatures (Table 1).

Nevertheless, δ-values of piezometer 32 showed statistically highest variation around the mean (Fig. 6), which is attributable to the influence of snowmelt that could only be detected for this piezometer (Fig. 5). As groundwater at the observed piezometers in the Vollnkirchener subcatchment is shallow (Orlowski et al., 2014), the snowmelt signal is allowed to move rapidly through the soil. Pulses of snowmelt water causing a depletion in spring and early summer was likewise observed by other studies (Darling, 2004; Kendall and McDonnell, 1998; Kortelainen and Karhu, 2004).

3.4 Isotopes of soil water

3.4.1 Spatial variability

Since soil water represents the interface between precipitation and groundwater, we did a snapshot soil sampling on four consecutive rainless days (1–4 November 2011) with additional information on distance to stream, TWI, land use, soil water content, soil texture, pH, and bulk density in the Vollnkirchener Bach subcatchment contributing to test hypothesis (2).

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Determining the impact of landscape characteristics on soil water isotopic signatures, we found no relationship between the above-mentioned soil parameters with the soil isotopic signatures in two depths (0.2 and 0.5 m), except for land use. This was potentially attributed to the small variation in soil textures (mainly clayey silts and loamy sandy silts), bulk densities, and pH values for both soil depths (Table 2). Water contents showed the greatest standard deviation within the two soil depths (Table 2), however exhibited no effect on soil water isotopes. Moreover, no tendency of higher TWI values with decreasing distance to stream was obvious. Garvelmann et al. (2012) found for a 0.9 km^2 humid catchment in the southern Black Forest (Germany) that soil profiles upslope or with a weak affinity for saturation (low TWIs) preserved the precipitation isotopic signal.

Generally, all soil water isotopic values fall on the local meteoric water line, indicating no evaporative enrichment of soil water (Fig. 8). The mean δ-values in the top 0.2 m of the soil profile is higher than further below, reflecting a stronger impact of precipitation in the topsoil (Table 2, Fig. 7). The δ-values of top soil and precipitation did not vary significantly statistically (Fig. 7), which is not the case for precipitation and subsoil. A mixing and homogenization of new and old soil water with depth could not clearly be seen in 0.5 m soil depth, which would have resulted in a lower SD (Song et al., 2011), but SDs of isotopic signatures in top and subsoil were similar (Table 2). Subsoil isotopic values were statistically equal to stream and groundwater isotopic values (Fig. 7) implying that the catchment was under baseflow conditions during the sampling campaign and that capillary rise of groundwater occurred. Nevertheless, the rainfall isotopic signal was not transferred through the soil to the groundwater body, resulting in the rejection of hypothesis (2). Similar observations were made by Garvelmann et al. (2012). The authors showed that groundwater was flowing through the soil in the riparian zone and dominated streamflow during baseflow conditions. Their comparison indicated that the percentage of pore water soil samples with a very similar stream water δ^2H signature is increasing towards the stream channel (Garvelmann et al., 2012). In contrast, we found no relationship between the distance to stream and soil isotopic values in the Vollnkirchener Bach subcatchment.

Comparing soil isotopic signatures between different land covers showed generally higher and statistically significantly different δ-values at 0.2 m soil depth under arable land as compared to forests and grasslands. However, all top soil isotopic values reflected precipitation isotopic signals (Fig. 8, top). For the lower 0.5 m of the soil column, isotopic signatures under all land use units showed statistically similar values; nevertheless, differing significantly from precipitation (Fig. 8, bottom).

Comparing soil water δ^2H values between top and subsoil under different land use units showed significant differences under arable and grassland but not under forested sites (Fig. 8, capital letters). This could be explained through the occurrence of vertical preferential flow paths and interconnected macro pore flow (in continuous root channels or earthworm burrows) (Buttle and McDonald, 2002) characteristic for forested soils (Alaoui et al., 2011). Alaoui et al. (2011) showed that macropore flow with high interaction with the surrounding soil matrix occurred in forest soils, while macropore flow with low to mixed interaction with the surrounding soil matrix dominates in grassland soils. The authors attributed the low efficiency of grassland soil macropores in transporting all water vertically downward to the fine and dense few topsoil layers caused by the land use that limit water flux into the underlying macropores. In general, the upper part of most agricultural human-impacted soils is restructured annually due to seasonal tilling, whereas the structure of forest soils, may remain unchanged for years and be uninterrupted throughout the entire soil profile (in particular the macropores and biopores) (Alaoui et al., 2011). Considering the bulk density in the Schwingbach catchment increasing values from forest (1.10 g cm^{-3}) over grassland (1.25 g cm^{-3}) to arable land soils (1.41 g cm^{-3}) were measured in the top soil. As reported in a study by Price et al. (2010) for North Carolina (USA), soils underlying forest trees generally feature low bulk density in a comparison with soils impacted by human land use. The reduced hydrological connectivity between top and subsoil under arable and grassland observed in the Vollnkirchener Bach subcatchment therefore led to different isotopic signatures (Fig. 8).

Although, vegetation cover has been proven to have an impact on soil water isotopes (Brodersen et al., 2000; Gat, 1996; Li et al., 2007), only few data are available for Central Europe (Darling, 2004). Burger and Seiler (1992) found that soil water isotopic enrichment under spruce forest in Upper Bavaria was double that beneath neighbouring arable land. However, in their study soil water isotopic signatures were not comparable to groundwater isotope values (Burger and Seiler, 1992). Brodersen et al. (2000) reported the effect of vegetation structure on $\delta^{18}O$ values of rainwater and soil water in the unsaturated zone in southern Germany. In their study, throughfall isotopic signatures of different tree species (spruce and beech) seemed to have a negligible effect on soil water isotopes, since soil water in the upper layers followed the seasonal trend in the precipitation input and had a very constant signature in greater depth. In contrast, Gehrels et al. (1998) detected slightly heavier isotopic signatures under forested sites at a field site in the Netherlands in comparison to non-forested sites (grassland and heathland), both showing isotopic signatures comparable to precipitation signals. For the Schwingbach catchment we conclude that the observed land use effect in the upper soil column is mainly attributed to different preservation and transmission of the precipitation input signal. It is most likely not attributed to distinguished throughfall isotopic signatures since top soil water isotopic signals followed the precipitation input signal under all land use units. The precipitation influence smoothed out with depth since soil water isotopes approached groundwater signatures at 0.5 m soil depth.

3.4.2 Seasonal isotope soil profiling

Examining the temporal effect of precipitation isotopic shifting in the soil, showed that isotope compositions of soil water varied seasonally (Fig. 9). Generally, more depleted soil water was found in the winter and spring (Fig. 9). Contrary, soil water was enriched in summer due to evaporation during warmer and drier periods (Darling, 2004). The depth to which soil water isotopes are significantly affected by evaporation is rarely more than 1–2 m below ground, and often less under temperate climates (Darling, 2004). For summer soil profiles in the Vollnkirchener subcatchment, no evidence for evaporation was obvious below 0.4 m soil depth. However, snowmelt isotopic signatures could be traced down to a soil depth of 0.9 m during spring rather than winter, pointing to a depth-translocation of meltwater in the soil, more remarkable for the deeper profile under arable land (Fig. 9, left panel). Furthermore, shallow soil water (< 0.4 m) showed larger SDs with values closer to mean seasonal precipitation inputs (Fig. 9). Winter profiles exhibited somewhat greater SDs in comparison to summer isotopic soil profiles, indicative for wetter soils and shorter residence times (Thomas et al., 2013). The observed seasonal amplitude smoothed out with depth as soil water isotope signals approached groundwater average. Generally, deeper soil water isotope values were relatively constant through time and space. Similar findings were made by Foerstel et al. (1991) on a sandy soil at Juelich, western Germany and by McConville et al. (2001) under predominately agriculturally used gley and till soils in Northern Ireland. Thomas et al. (2013) likewise observed that soil water isotope samples from shallow soils (≤ 30 cm) were comparable to precipitation isotopic composition, while samples from intermediate soils (40–100 cm) plot near the groundwater average for a forested catchment located in central Pennsylvania, USA. Furthermore, Tang and Feng (2001) showed for a sandy loam soil sampling site in New Hampshire (USA) that the influence of summer precipitation decreased with increasing depth, and soil at 0.5 m can only receive water from large storms. For summer soil profiles under arable land, precipitation input signals likewise decreased with depth (Fig. 9, left panel).

Generally, the replacement of old soil water with new infiltrating water is dependent on the frequency and intensity of precipitation and the soil texture, structure, wetness, and water potential of the soil (Li et al., 2007; Tang and Feng, 2001). It is usually more efficient in a wet year than in a dry year (Tang and Feng, 2001). As a result of soil water recharge near the surface, the amount of percolating water decreases with depth and consequently, deeper soil layers have less chance to obtain new water (Tang and Feng, 2001). Furthermore, in the growing season, the percolation depth is additionally limited by plants' transpiration (Tang and Feng, 2001). For the Schwingbach catchment we conclude that the influence of new percolating soil water decreased with depth as no remarkable seasonality in soil isotopic signatures was obvious at > 0.9 m and constant values were observed through space and time.

3.5 Local Meteoric Water Line and isotopic comparison of water cycle components

The linear relationship of δ^2H and δ^{18}O content in local precipitation, results in a local meteoric water line (LMWL) (Fig. 10), which can be utilised to link the relative contribution of seasonal precipitation to ground and surface water sources (Wassenaar et al., 2011). The global meteoric water line (GMWL) established by Craig (1961a), and more recently refined by Rozanski et al. (1993) is $\delta^2\mathrm{H} = 8.13 \times \delta^{18}\mathrm{O} + 10.8$ ‰. It provides a valuable benchmark against which regional or local waters can be compared (Song et al., 2011). The slope of the LMWL of the Schwingbach catchment is well in agreement with the one from the closest GNIP station in Koblenz ($\delta^2\mathrm{H} = 7.67 \times \delta^{18}\mathrm{O} + 2.48$ ‰; $R^2 = 0.98$), but is slightly lower in comparison to the revised GMWL, showing stronger local evaporation conditions. Since evaporation causes a differential increase in δ^2H and δ^{18}O values of the remaining water, the slope for the linear relationship between δ^2H and δ^{18}O is lower in comparison to the GMWL (Rozanski et al., 2001; Wu et al., 2012). The lower intercept (d-excess), dependent on the humidity and temperature conditions in the evaporation region (Mook, 2001), nevertheless, shows that moisture recycling did obviously not play a major role in the study area.

Considering isotope samples of the different water cycle components in comparison with the LMWL revealed that mean isotope values of snow samples were for δ^2H approximately 84 ‰ lighter that mean precipitation isotopic signatures (Fig. 10). Stream water isotope samples of both creeks (Schwingbach and Vollnkirchener Bach) fell on the LMWL, showing slight evaporative enrichment for few samples (Fig. 10). Moreover,

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isotopic values for stream water were almost identical to those found in groundwater (Table 1, Fig. 10).

Same observations were made by Jin et al. (2010) for the Red Canyon Creek watershed (Wyoming, USA), indicating good hydraulic connection between surface water and shallow groundwater and by Klaus et al. (2014) for a low-mountainous forested watershed in South Carolina (USA), comparable to the Schwingbach catchment. Furthermore, isotopic similarities between stream and groundwater pointed out that surface water was mainly replenished by the groundwater, except for extreme storm events (Orlowski et al., 2014). Same was observed by Zhang et al. (2013) for the Tarim River Basin in China. However, in the Vollnkirchener subcatchment arable land groundwater isotopes were slightly heavier and hydrologically decoupled from the Vollnkirchener Bach.

4 Conclusions

Conducting a stable water isotope study in the Schwingbach catchment helped to identify relationships between precipitation, stream, soil, and groundwater in a developed catchment. The close isotopic link between groundwater and the streams revealed that groundwater controls streamflow. Moreover, it could be shown that groundwater was predominately recharged during winter but was decoupled from the annual precipitation cycle. Even so streamflow and groundwater head levels rapidly responded to precipitation input, there was no evidence for a larger contribution of precipitation to both water cycle components. This was underlined by the fact that no remarkable seasonality in soil isotopic signatures as interface between precipitation and groundwater was obvious at > 0.9 m and constant values were observed through space and time.

Nevertheless, the lack of temporal variation in stable isotope time series of stream and groundwater (with few exceptions) limited the application of classical methods of isotope hydrology (mean transit time estimation, hydrograph separation) in the Schwingbach catchment. Still, our dual isotope approach was valuable for determin-

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ing the connectivity and disconnectivity between different water cycle components. Together with results of a former hydrometric study in the same catchment (Orlowski et al., 2014), knowledge about water cycle component interactions will be utilised to develop catchment specific process-based hydrological models. Since groundwater seems to be the major driver of streamflow, a groundwater dating campaign is planned in the future.

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Table 1. Descriptive statistics of δ^2H and δ^{18}O in precipitation, stream-, and groundwater over the two-year observation period including all sampling points.

Sample type	Min		Max		Mean		± SD		N
	δ^2H [‰]	δ^{18}O [‰]	δ^2H [‰]	δ^{18}O [‰]	δ^2H [‰]	δ^{18}O [‰]	δ^2H [‰]	δ^{18}O [‰]	
Precipitation	−167.6	−22.4	−8.3	−1.2	−43.9	−6.2	23.4	3.1	592
Vollnkirchener Bach	−66.3	−10.0	−26.9	−6.7	−58.0	−8.4	2.8	0.4	332
Schwingbach	−139.7	−18.3	−47.2	−5.9	−58.2	−8.4	4.3	0.6	463
Groundwater meadow	−64.9	−9.2	−50.8	−5.7	−57.6	−8.2	1.6	0.4	375
Groundwater arable land	−91.6	−12.3	−49.5	−6.8	−56.2	−8.0	3.7	0.5	338
Groundwater along stream	−94.5	−13.0	−49.5	−7.0	−59.9	−8.5	6.8	0.9	108

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Table 2. Mean and SD for soil physical properties and isotopic signatures in 0.2 and 0.5 m soil depth (N = 52 per depth).

	δ^2H [‰]		$\delta^{18}O$ [‰]		water content [% w/w]		pH		bulk density [$g\,cm^{-3}$]	
	0.2 m	0.5 m	0.2 m	0.5 m	0.2 m	0.5 m	0.2 m	0.5 m	0.2 m	0.5 m
Mean	−46.9	−58.5	−6.6	−8.2	16.8	16.1	5.0	5.3	1.3	1.3
± SD	8.4	8.3	1.2	1.2	7.2	8.3	1.0	1.0	0.2	0.2

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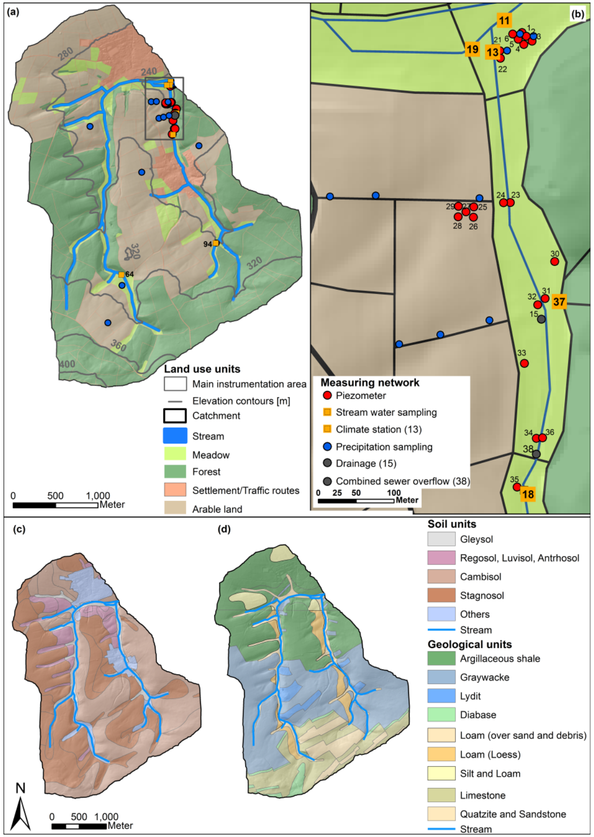

Figure 1. Maps of **(a)** land use and instrumentation, **(b)** main monitoring area, **(c)** soils, and **(d)** geology.

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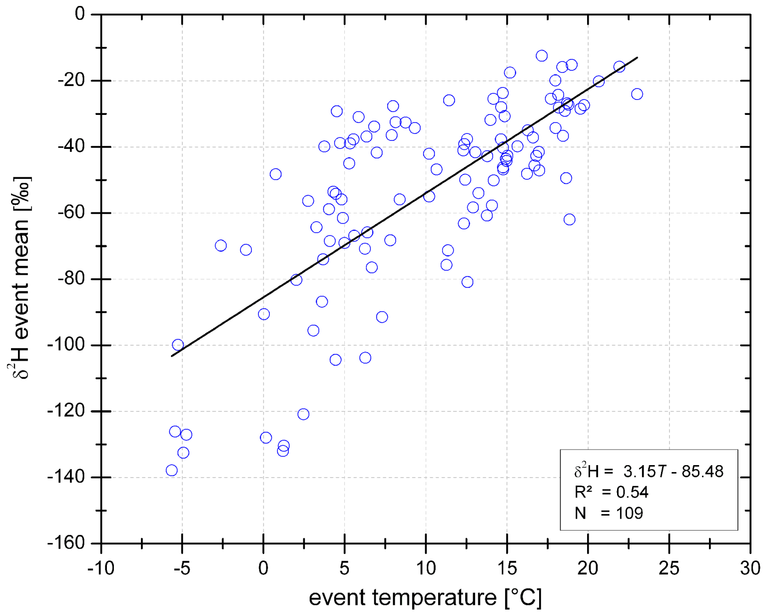

Figure 2. Relationship between event mean δ^2H values in precipitation and air temperature.

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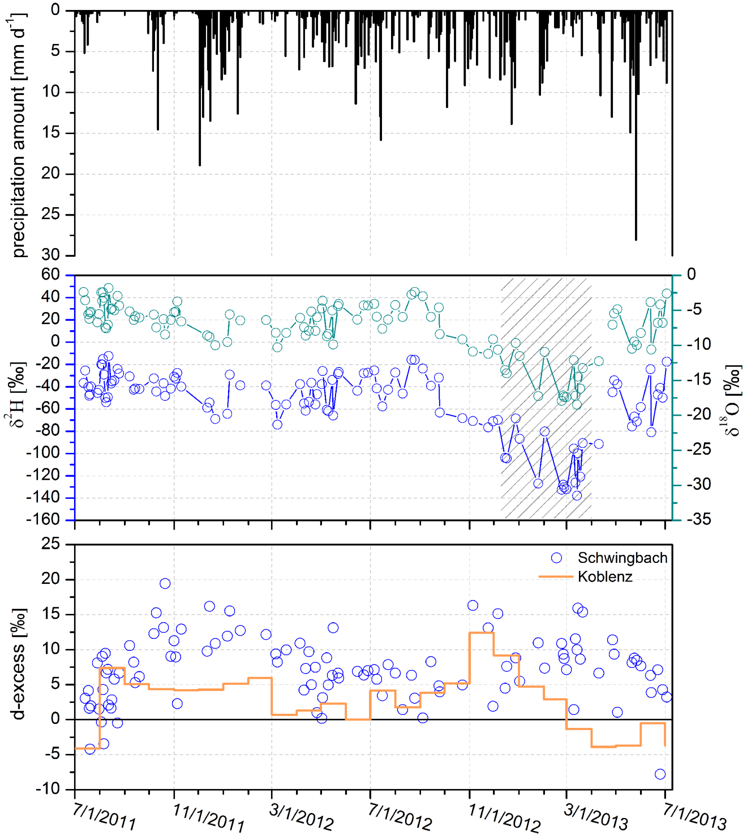

Figure 3. Temporal variation of precipitation amount, isotopic signatures (δ^2H and δ^{18}O) including snow samples (grey striped box), and d-excess values for the study area compared to monthly d-excess values (July 2003–July 2005) of GNIP station Koblenz with reference d-excess of GMWL (d = 10; dashed line).

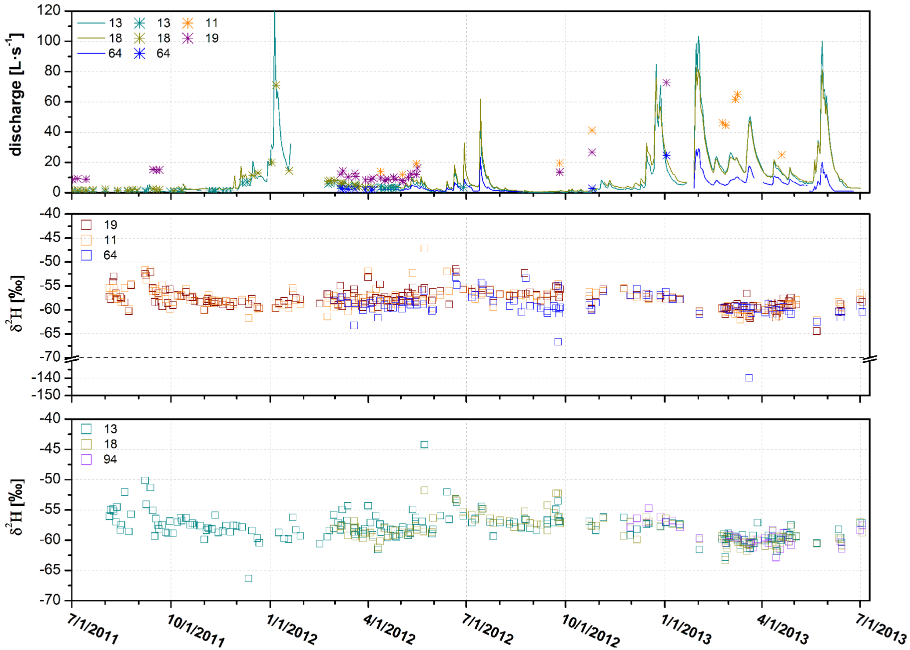

Figure 4. Mean daily discharge at the Vollnkirchener Bach (13, 18) and Schwingbach (site 11, 19, and 64) with automatically recorded data (solid lines) and manual discharge measurements (asterisks), temporal variation of δ^2H of stream water in the Schwingbach (site 11, 19, and 64) and Vollnkirchener Bach (site 13, 18, and 94).

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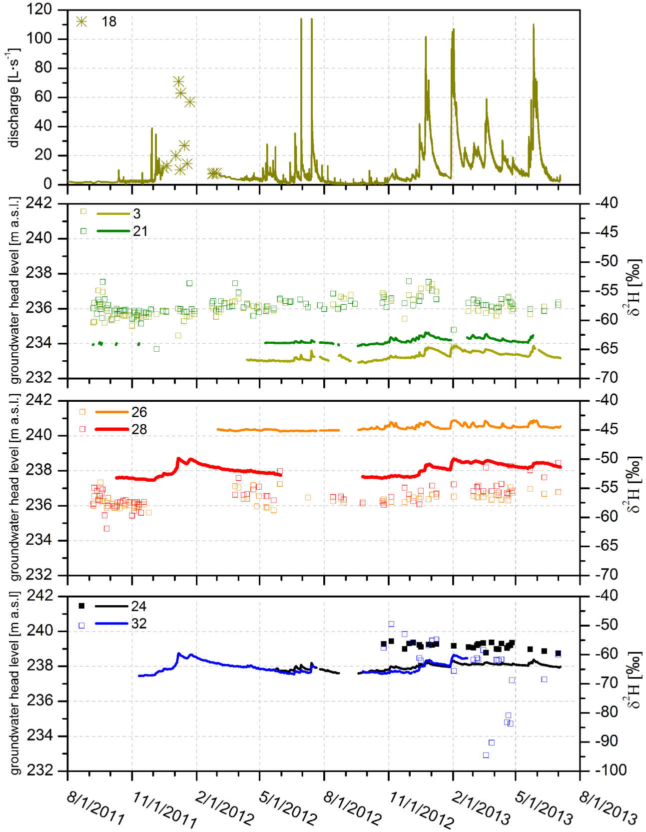

Figure 5. Temporal variation of discharge at the Vollnkirchener Bach (site 18), groundwater head levels, and δ^2H values (coloured dots) for selected piezometers under meadow (site 3 and 21), arable land (site 26 and 28), and beside the Vollnkirchener Bach (site 24 and 32).

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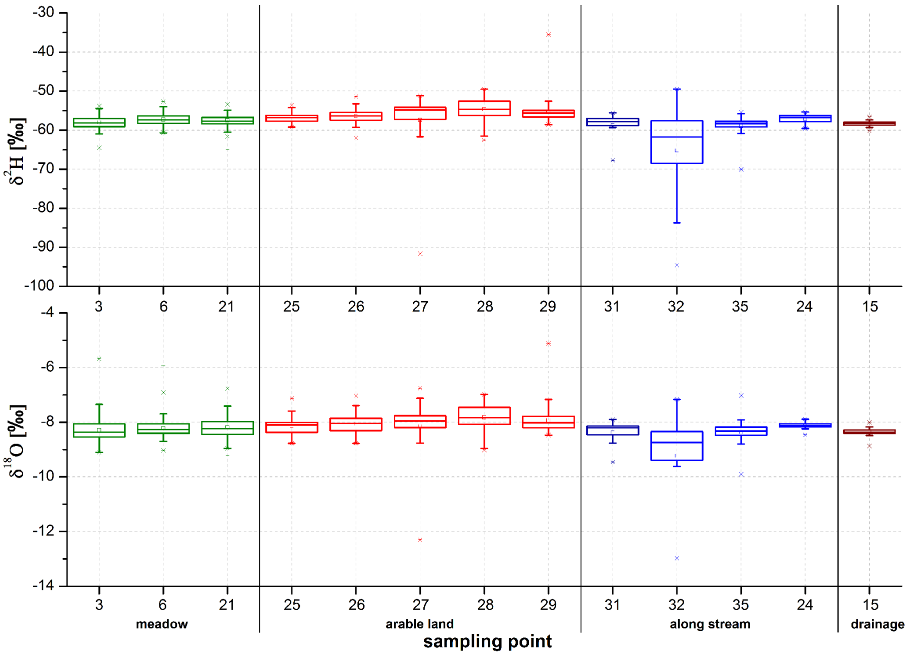

Figure 6. Boxplots of isotopic variation (δ^2H and δ^{18}O) in groundwater under meadow (site 3, 6, and 21), arable land (site 25–29), and along the stream (site 31, 32, 35, and 24) as well as for a drainage (site 15).

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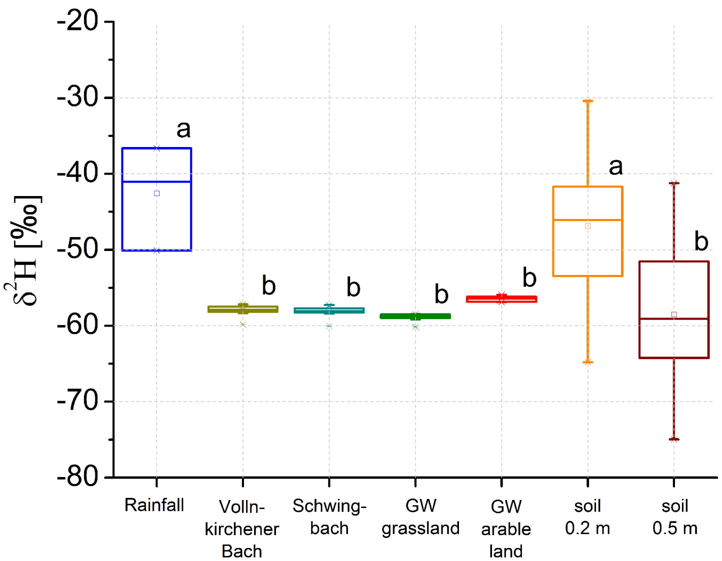

Figure 7. Boxplots of δ^2H values comparing rainfall, stream, groundwater, and soil isotopic composition in 0.2 and 0.5 m depth ($N = 52$ per depth). Different letters indicate significant differences ($p \leq 0.05$).

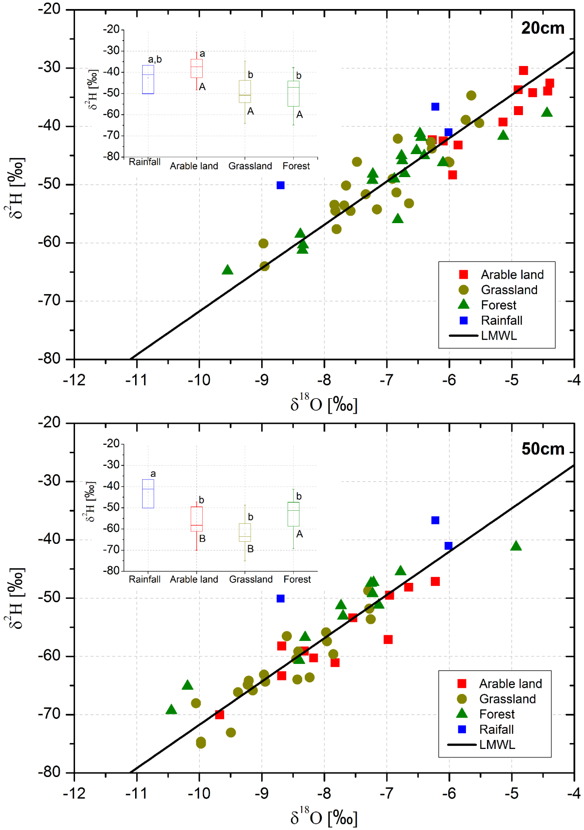

Figure 8. Dual isotope plot of soil water isotopic signatures in 0.2 and 0.5 m depth compared by land use including rainfall isotope data from 19, 21, and 28 October 2011. Insets: boxplots comparing δ^2H isotopic signatures between different land use units and rainfall (small letters) in top and subsoil (capital letters). Different letters indicate significant differences ($p \leq 0.05$).

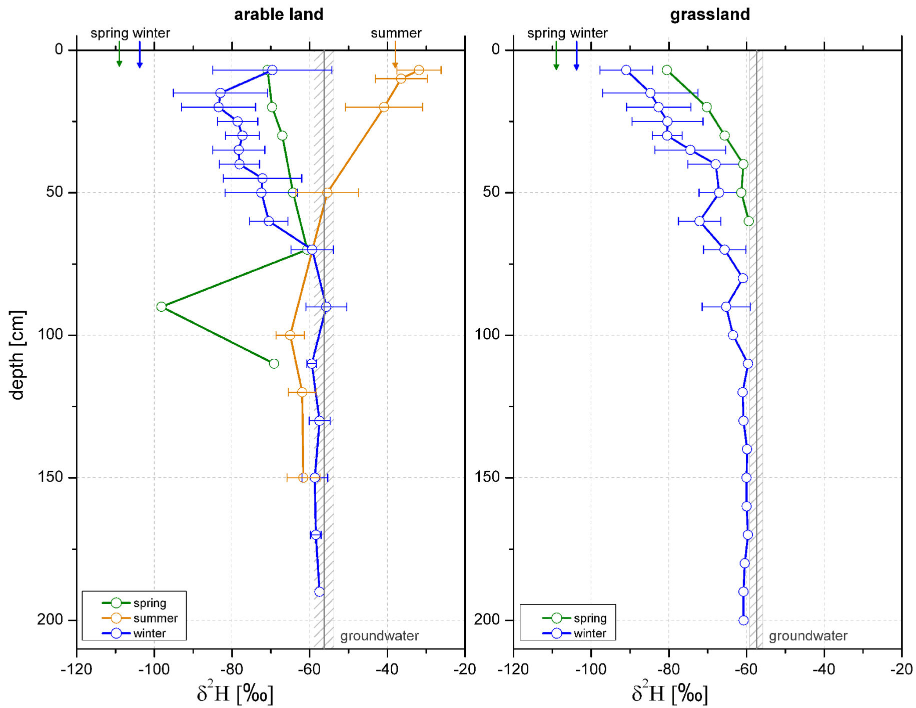

Figure 9. Seasonal δ^2H profiles of soil water for winter, summer, and spring based on soil samplings conducted on 28 August 2011 (summer), 28 March 2013 (winter), and 14 April 2013 (spring). For reference, mean groundwater (grey shaded) and mean seasonal precipitation δ^2H values are shown (coloured arrows at the top).

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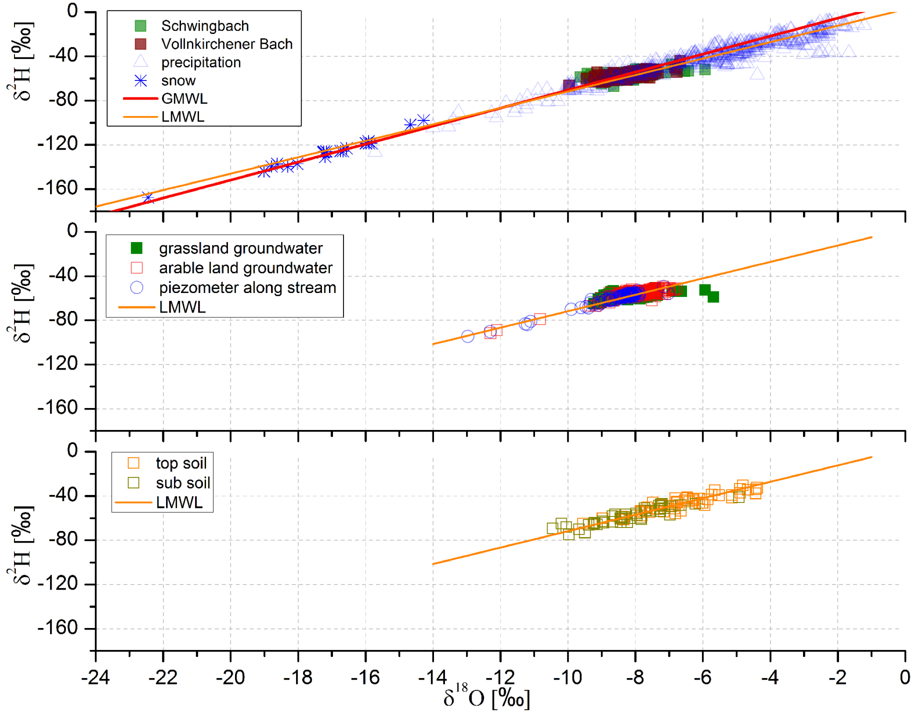

Figure 10. Local Meteoric Water Line for the Schwingbach catchment (LMWL) in comparison to GMWL, including comparisons between precipitation, stream water, groundwater, and soil water isotopic signatures.

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