Hydrological Processes in the Subsurface Investigated by Water Isotopes and Silica

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Abstract: The hillslope rainfall-outflow interactions, groundwater fluxes, and hydrological balance were examined in the small mountainous headwater catchment Uhlířská, the Jizera Mountains, the Czech Republic. The hillslope soil profile is formed by paleozoic crystalline bedrock overlaid by shallow highly permeable shallow Cambisol, and by thick saturated glacial deposits in the valley, overlaid by Histosol. A quick communication of the vadose zone with the granitic bedrock via preferential subsurface flowpaths is hypothesised, in agreement with the observation of storm-caused instant water transformation to outflow through the permeable Cambisol. A quick response of a high magnitude outflow occurs regularly, although the surface runoff is very rare. Standard climatic and hydrological monitoring in the Uhlířská catchment is supplemented by the measurements of the soil moisture, soil pore water suction, subsurface hillslope stormflow in the vadose zone, and water table fluctuation in the saturated subsurface, and is accompanied by water sampling for the analyses of the contents of the isotope ^{18}O and ^{2}H and geochemical tracer silica in the form of SiO_2. The episode based isotopic data serve for the separation of the outflow hydrograph to determine the contributions of the event and pre-event water in the hypodermic hillslope outflow and in the catchment outflow. The variation of silica content in the water cycle components was examined to assess the contributions from the soil profile and the aquifer. Up to 75% of the event catchment runoff was assigned to pre-event water, of which about 50% had been stored in the shallow soil subsurface on the hillslopes. The hypothesis was confirmed that the hillslope soil layers control the distribution of the flow into the groundwater recharge and/or the shallow subsurface flow during the rainfall-runoff episode.

Keywords: rainfall to runoff response; isotopes; geochemical tracers; streamflow generation; subsurface stormflow; groundwater recharge

Many experiments, models, and concepts of the streamflow generation in headwater catchments have been adopted within the past decades, summarised in a structured framework of key hypotheses on where the water goes when it rains, which pathways it takes to the stream and how long it resides in the catchment. Among the first concepts, Horton (1933) developed a theory of excess effective rainfall and overland flow generating major amounts of the flood runoff. Research

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work in the 1950's and 1960's resulted in concepts of subsurface runoff sources, such as "translatory flow" (Hewlett & Hibbert 1965) or subsurface stormflow (Whipkey 1965), synthesised and further developed e.g., by Dunne and Black (1970). Although this type of hydrological response can be seemingly well formalised by a simple or more complicated inflow-outflow relationships (Beven 1997), the introduction of geochemical and isotopic tracing since the 1960's has shown that these relatively simple concepts are not satisfactory to reveal the causalities of the runoff formation (McDonnell 2003). The study of the water dynamics by means of natural tracers has been typically targeted to employ the effects of stable isotopes of oxygen ^{18}O and hydrogen ^{2}H (Kendall & McDonnell 1998) and elements such as silica as SiO_2 (Kennedy 1971) or combined with Ca^{2+} (Iorgulescu *et al.* 2007). Such investigation increases the quality of the present research fundamentally by adding an independent set of information on the catchment outflow generation.

While the first isotopic and geochemical studies in small temperate headwaters introduced a novel concept of the dominance of the pre-event ("old") water during runoff events, the crucial contribution of the more recent tracer studies in headwaters since the 1980's (e.g., DeWalle *et al.* 1988; Buzek *et al.* 1995; Holko 1995; Rodhe *et al.* 1996; Soulsby *et al.* 2000; Burns *et al.* 2001; Katsuyama *et al.* 2001; Vitvar *et al.* 2002; Asano *et al.* 2002; Uhlenbrook *et al.* 2002; Weiler *et al.* 2003; Maloszewski & Zuber 1996) lies in the fact that they highlighted various conceptual paradoxes which would not have been easily addressed without the use of tracers. On one hand, it is known that the water residing in a small catchment prior to the rainfall ("old" or "pre-event" water) is replaced by the event water throughout the porous zone and discharged to the stream, although the mechanics of the subsurface pressure transformation is not yet completely understood. On the other hand, however, a contradiction is found between the determination of these basic streamflow components using formally identical approaches of isotopes (^{18}O or ^{2}H) and elements such as silica or calcium, showing that the water of the same origin or residence time may often move via independent geochemically marked pools/flowpaths and *vice versa*. When addressing these paradoxes, Kirchner (2003) states that there are several pools (or a continuum of stores) of pre-event water within a catchment which are mobilised under different conditions and discharged through variable pathways, which cannot be assumed to be spatially homogeneous and whose hydrological response cannot be considered linear either. In addition, the subsurface topography often differs significantly from the surface topography. Understanding of this complex process leads to current novel approaches toward the assessment of hillslopes as elementary units essential for understanding the catchment response as a whole system. These approaches have been applied in several experimental catchments (Freer *et al.* 2002; Seibert *et al.* 2003; McDonnell *et al.* 2006; Tromp-van Meerveld & McDonnell 2006), and two-area (Uchida *et al.* 2006) or multi-area (SLope InterComparison Experiments SLICE; Retter *et al.* 2006) comparisons. In addition, many of the novel findings about water fluxes in subcatchments or hillslopes (residence times, pre-event water contribution) serve as parameters in the rainfall-runoff models, often supplying an alternative model evaluation besides the quality of the runoff simulation fit (Seibert & McDonnell 2002).

The small headwater experimental catchment Uhlířská has been operated by the Czech Hydrometeorological Institute since early 1980's, originally aimed at studying the extreme outflow situations (Blazkova & Beven 1997) caused by rainfalls in heavily deforested regions due to the atmospheric pollution. This is nowadays complemented with studies regarding the water balance or quality of drinking water supplies. The flow mechanisms transforming rainfall into runoff in both the variably saturated soil profile on the instrumented hillslope and within the entire catchment alike have been studied since late 1990's (Šanda *et al.* 2004).

More recent studies highlighted the processes of infiltration and runoff formation in deforested soils, storage potential, and residence time of water in the subsurface (Šanda *et al.* 2007a). The crucial questions to be addressed in the ongoing research include the assessment of the pre-event runoff component in terms of the subsurface pool type (unsaturated zone or aquifer) and isotopical and chemical evidence of pre-event water displacement and transport to the stream. Based on the first estimations of the soil and groundwater residence times (Šanda *et al.* 2009), we hypothesise that the soil water pool plays a dominant role in the streamflow generation, supplying both short and long term flow components.

MATERIALS AND METHODS

The Uhlířská catchment (1.78 km^2), is situated in the humid temperate mountainous region of the Jizera Mountains, northern Czech Republic, in the region of Nisa and Labe headwaters. The average air temperature is 4.6°C, average annual precipitation is 1400 mm, annual average discharge is 63 l/s (1117 mm) (Bercha *et al.* 2008). Based on this information, the annual average loss of water is 283 mm. The Uhlířská is a typical catchment with the crystalline bedrock forming Cambisols as does 60% of the area of the Czech Basin, soils are typically shallow and highly permeable with preferential pathways. The streamflow generated by storms can be therefore of a quick response and high magnitude (Šanda *et al.* 2006).

Focusing on the flow processes in the subsurface, the aim of the hydrological research is to reveal the flow mechanism transforming rainfall into runoff in both the variably saturated soil profile on the instrumented hillslope and within the scope of the catchment. Both of the natural elements (^{18}O and silica) had been sampled at selected sites in the catchment since the spring of 2006 (Šanda *et al.* 2007b). These activities covered the sample collection of precipitation, snowmelt, snowcover, subsurface stormflow, groundwater, soil water from soil suction cups, and the stream outflow at two gauging stations – for the whole catchment UHL and subcatchment POR (Figure 1). The samples for ^{18}O analyses were collected in liquid rainfall daily, snowfall weekly, snowcover weekly, soil subsurface stormflow at the subsurface trench on the hillslope (episode based each 6 hours), snowmelt (event based daily), streamflow at two gauging profiles (daily or episode based each 6 hours), soil pore water (4 locations, 2 horizons) monthly and shallow groundwater (4 locations) monthly. The analyses of the samples for ^{18}O concentration were performed in the Czech Geological Survey in Prague using Finnigan Mat 250 mass spectrometer. Some of the samples were cross-checked on the Liquid Isotope Analyzer, LGR laser spectrometer, Slovak Geological Survey of Dionyz Stur, Bratislava, for both ^{18}O and ^{2}H. Additionally, ^{18}O and ^{2}H are sampled in the cumulated monthly precipitations and the stream outflow is acquired monthly and analysed in the IAEA Isotope Hydrology laboratory within the framework of GNIP and GNIR programmes of IAEA (IAEA 2006; Vitvar *et al.* 2007), using the Finnigan MAT DeltaPlus mass spectrometer. All ^{18}O values are expressed as δ^{18}O, and all ^{2}H values are expressed as δ^{18}H in per-mil of the Vienna Standard Mean Ocean Water, with precision of $\delta\,^{18}$O ± 0.1 per mil V-SMOW (± 0.2 per mil V-SMOW for laser spectroscope) and $\delta\,^2$H ± 1 per mil V-SMOW for both mass spectrometry and laser spectroscopy.

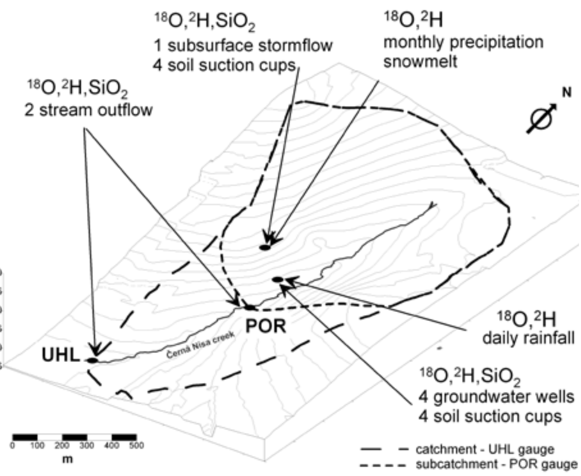

Figure 1. Water sampling locations in the experimental catchment Uhlířská

The analyses for silica were performed in the Czech Geological Survey in Prague, using ICP-OES Thermo Jarrell Ash, Iris Advantage device. The values are expressed as SiO_2 concentration in water in mg/l with precision of ± 0.1% of the measured value.

Besides the most recent activities in the area of the environmental hydrological isotopes and geochemical tracers, detailed long-term automated measurements of climatic and hydrological components have been performed within the last decade. The measurements comprise precipitation, air and surface temperature, net radiation, air humidity, barometric pressure and wind speed measurements. Soil suction, soil moisture, and groundwater level at the selected locations are also acquired on continuous basis. In the subsurface, the stormflow is collected for the outflow measurements on the hillslope. The setup, built for the subsurface stormflow observation, is recently utilised for the isotope and silica contents sampling as well (Šanda *et al.* 2007a). The site is accompanied with a basic climatic station recording the air temperature, net radiation, wind speed, and humidity on a continuous basis (Šanda *et al.* 2004).

Chemical separation of the runoff components during an episodic event (Buttle 1994) is based on the following set of Eqs (1)–(3) for the outflow intensity and isotope contents in the water.

$$Q_t = Q_s + Q_n \quad (1)$$

$$c_t Q_t = c_s Q_s + c_n Q_n \quad (2)$$

$$R_s = \frac{Q_s}{Q_t} = \frac{c_t - c_n}{c_s - c_n} \quad (3)$$

where:

Q_t – total outlow (m^3/s)

Q_s – amout of pre-event water in the outflow (m^3/s)

Q_n – amout of the event water in the outflow (m^3/s)

c_s – concentration of the tracer in the streamwater prior to the event ($\delta^{18}O$)

c_n – concentration of the tracer in the streamwater during the event ($\delta\,^{18}O$)

R_s – instantaneous fraction of the pre-event water in the outflow during the event (–)

For the purposes of this paper, one rainfall-outflow event that occurred during 21–23. 6. 2006 was selected to demonstrate the approach. The flow in the stream is considered episodic if the instant flow rate exceeds 75% probability of exceedance of the mean daily discharges at the rising limb and decreases below the same value on the falling limb of the hydrograph. The sampling setup for the event included daily totalled rainfall sample and the subsurface outflow and stream outflow (when available) discrete samples once a day for this episode. The values obtained of $\delta\,^{18}O$ in the rain and outflow were linearly interpolated in time. For the outflow samples, the exact time of sampling was used for interpolation, whereas the medium interval time (12:00) was used as a reference value for the rain sample. The values of $\delta\,^{18}O$ in the rain and flow were calculated for intervals of 10 minutes in the available flow records to enable the calculations according to Eqs (1)–(3).

RESULTS AND DISCUSSION

The streamflow intensity of the entire catchment outflow is shown in Figure 2. For the non-vegetation season, the ^{18}O measured (Figure 3) indicates the replacement of pre-melt water in the subsurface outflow and stream outflow during the snowmelt seasons. The outflow is transformed by the variably saturated soil profile and then in the saturated aquifer. A quick response of the soil profile to the change in $\delta^{18}O$ is evident during storms (Figure 3). It supports the hypothesis of the partial transformation of the rainfall into runoff within the soil profile, employing the preferential pathways. With the time, the differences in $\delta\,^{18}O$ found between the subsurface stormflow and the catchment streamflow diminish, probably due to the extreme nature of a storm event where most of the pre-event water is already displaced with the causal rainwater.

The development of the concentration values of SiO_2 at the same sampling sites is similar (Figure 4). A significant dilution of silica by storms in the outflow from the soil profile and the whole catchment is observed similar to the findings by Scanlon *et al.* (2001). The variations of SiO_2 prior to the snowmelt show a range of low concentrations, however, a rapid rise is observed since the commencement of the snowmelt. Silica precipitation or decreased dissolution at low temperatures is the most probable cause. Also, the pre-event baseflow drains the aquifer via per-

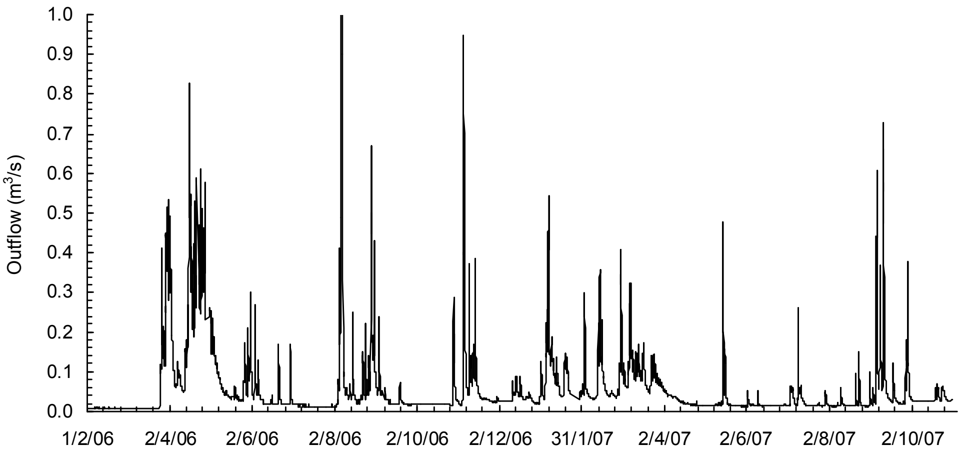

Figure 2. Daily Outflow hydrograph at the stream gauging profile Uhlířská (UHL)

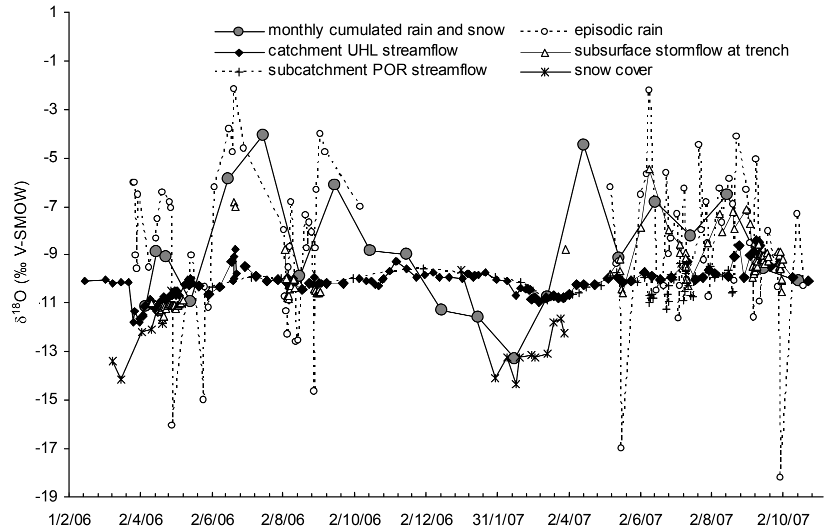

Figure 3. Isotope $\delta^{18}O$ content in rain, snow, subsurface outflow and streamflow in the Uhlířská catchment

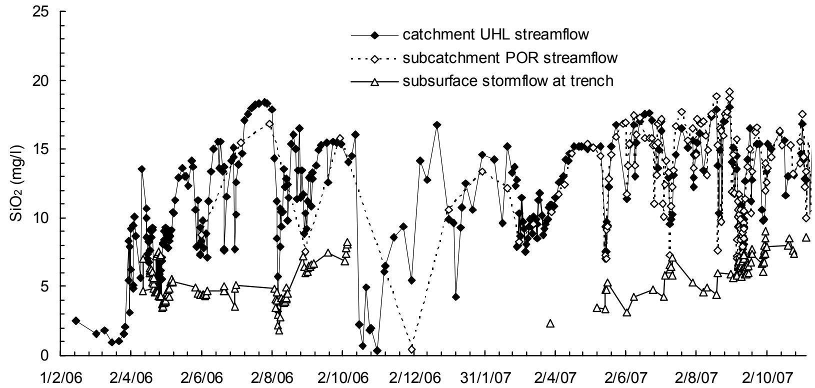

Figure 4. Concentration of silica (SiO_2) in subsurface outflow and streamflow in the Uhlířská catchment

manently washed pathways, where silica can not be dissolved in water to higher concentrations.

On the contrary, the snowmelt water replaces the soil water stagnant during the winter season in the soil profile. If a sufficient time is available to dissolve silica in the soil water, the soil water significantly contributes to its rapid rise in the streamflow. The content of SiO_2 exhibits observable outflow-related variations during the summer. The subsurface water, such as soil pore water and groundwater in the saturated zone, shows a generally progressive increase in SiO_2 with increasing depth (Figure 7), suggesting that the weathering dominates over the biological cycle of silica. The values of SiO_2 at 30 cm depth in Histosols are comparable to those in average streamwater, supporting the hypothesis of the dominant role of the soil horizon in the streamflow generation, as verified by ^{18}O. This SiO_2 is likely to be biogenic (Derry *et al.* 2005). The rapid decrease of silica during the time of the base flow in October, 2006 may imply the fact that silica was depleted by diatoms in bloom after the extremely hot summer and autumn of 2006.

According to the ^{18}O values in the subsurface porous space of the catchment, the soil profile showed the dominant role in the initial mixing of the rainfall water, resulting furthermore in an almost constant content of ^{18}O in the groundwater (Figure 5). The fluctuation of ^{18}O in the subsurface was of much narrower amplitude compared with the amplitude in the rainfall (Figures 5–6). Moreover, the variations of the silica content in the soil pore water and groundwater were much less pronounced than in the catchment outflow except the borehole PST in the riparian discharge zone (Figure 7).

Figure 8a provides a comparison of $\delta^{18}O$ and δ^2H in monthly accumulated precipitations and monthly streamwater samples for the period of May 2006–October 2007 in the form of the meteoric water line and outflow water lines. In analogy,

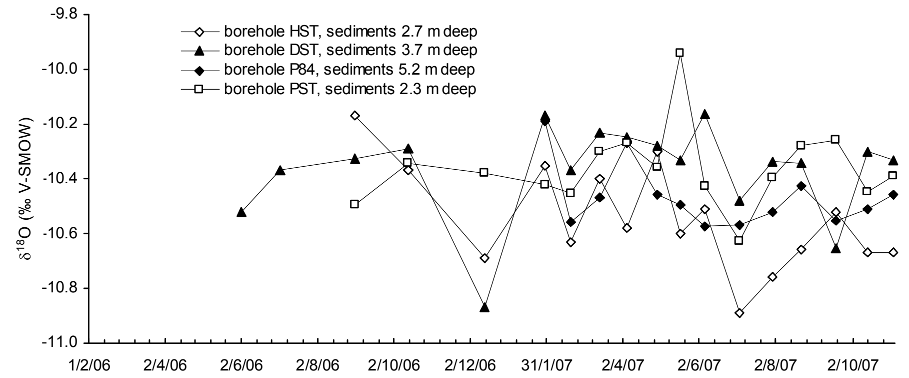

Figure 5. Isotope $\delta^{18}O$ content in groundwater in four boreholes in the Uhlířská catchment

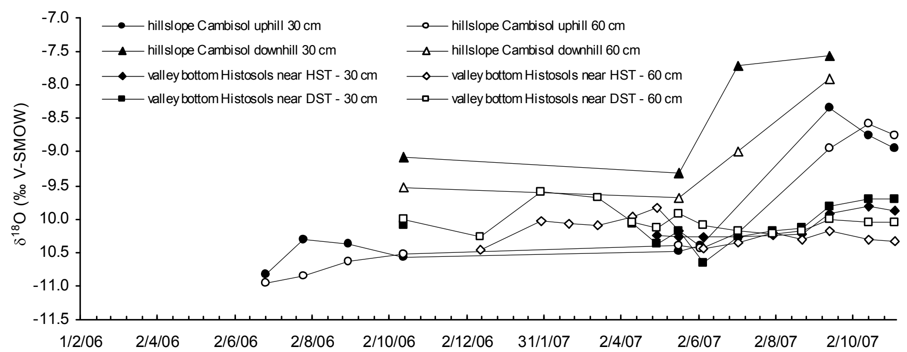

Figure 6. Isotope $\delta^{18}O$ in soil matrix pore water at the hillslope and valley sampling sites in the Uhlířská catchment

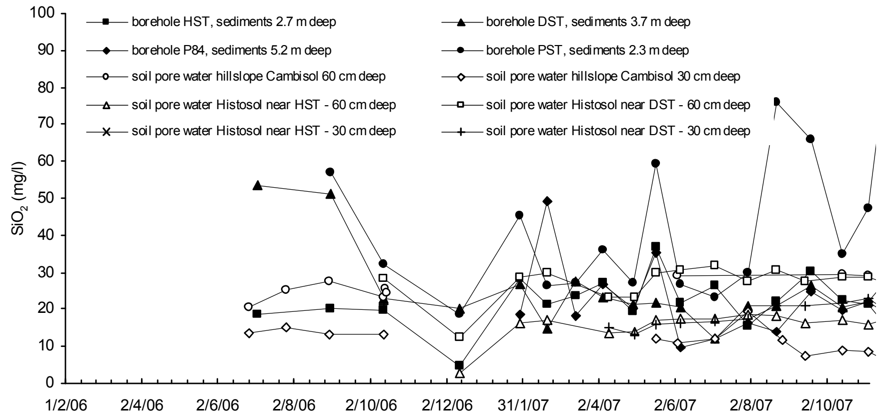

Figure 7. Silica (SiO_2) in soil matrix pore water and groundwater in the Uhlířská catchment

Figure 8b shows daily or hourly data for the rainfall runoff episodes during the vegetation season of 2007 as compared with the long term pore water and groundwater components. There is evidence of a shallow pre-event water found in the outflow as plotted in Figure 8b below the meteoric water line based on daily or more detailed data. These components indicate a near surface isotopic fractionation induced by evaporation.

The development of the isotope ^{18}O and silica content in water for the stormflow episode of 21–23. 6. 2006 is shown in Figures 11–12. Figure 9 shows the separation of the outlet gauge hydrograph based on the Eq. (3), considering the pre-event content in the stream outflow of $\delta^{18}O = -10.1$‰ (measured in the outflow prior to the event). An analogical separation was performed for the experimental trench by means of soil suction cups with the pre-event water content in soil pores of $\delta\,^{18}O = -10.9$‰ (Figure 10) The separation of the catchment hydrograph shows a significant pre-event water component (not less than 75%), whereas the subsurface outflow from the shallow soil profile shows that 46–51% of the subsurface

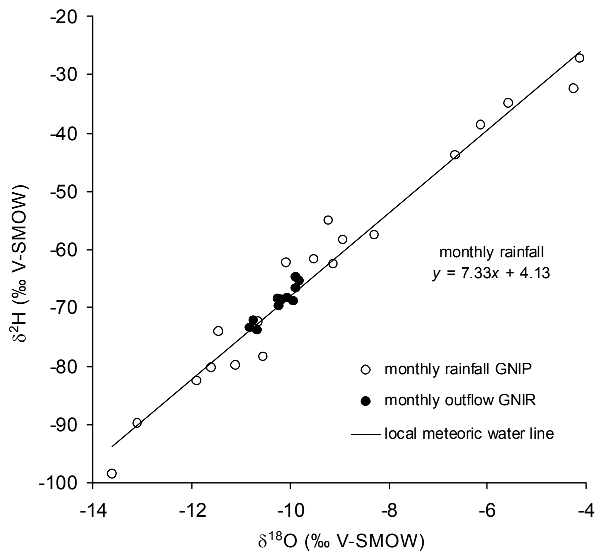

Figure 8a. Relationship of monthly $\delta^{18}O$ and $\delta^{2}H$ values (May 2006–October 2007) in rainfall and streamflow along the Local Meteoric Water Line, Uhlířská catchment

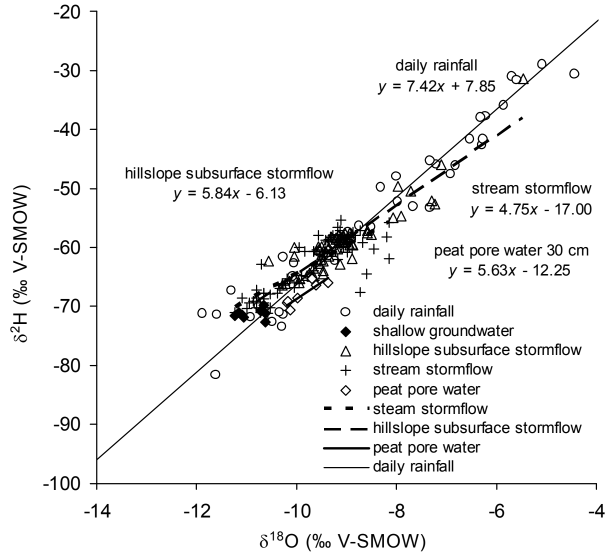

Figure 8b. Water lines – relationships of $\delta^{18}O$ and $\delta^{2}H$ for storm events in the vegetation season of 2007, Uhlířská catchment

storm flow was composed of the pre-event water. These findings indicate an important role of the soil profile and the valley aquifer for the runoff generation and isotopic composition of storm outflow.

The observation of the robust buffering effect of the porous space of the catchment, i.e. no significant change in the isotopic values in the outflow according to the significantly deviated isotopic signal from the mean in precipitation, is in contradiction with the high dilution of silica during the same rain event if the peakflow and baseflow concentrations of silica are compared. Based on random analyses, rainwater is assumed to contain no silica. While the values of silica in the baseflow ranged between 12–20 mg/l, the peak concentrations dropped to values very similar to those observed in the subsurface stormflow from the soil profile. Using Eqs (1)–(3) with the event water concentration c_n equal zero, the contribution of the groundwater to the stream peakflow would be much lower than in the case of isotopic separation. The isotopic compositions of the groundwater in the valley and the soil pore water on the hillslopes are practically indistinguishable (Figures 5 and 6), the silica content doubled or even tripled in the groundwater, based on the comparison of the silica contents in the Histosol and Cambisol throughout the year.

These findings strongly support the hypothesis that the major displaced component during the stormflow is the shallow soil pore water, drained via preferential pathways, rather than the displaced shallow groundwater. Other explanation can be found through dynamically changing water chemistry during the event while relatively stagnant soil pore water and groundwater are displaced and actuated. Depending on the type of the precipitation event, the ratios, i.e. the contributions towards the outflow provided by the subsurface water pools (i.e. soil pore water and groundwater) vary between the events and within an event as well.

CONCLUSIONS

This paper combines the analysis of the fluctuations of silica and of ^{18}O, ^{2}H in rain, subsurface stormflow, streamflow, soil pore water and groundwater with the use of isotopic and chemical separation techniques and the observation of temporal variations of water isotopes contents and its chemistry. The combined use of both approaches provided a deeper insight into the processes of runoff generation and rainfall-runoff transformation within the catchment subsurface. The hypothesis of an important role of the soil profile on the lateral hillslope stormflow and groundwater recharge was confirmed. This quick subsurface mixing is assumed to be the principal runoff generation process in the catchment. Streamwater with constant isotopic and geochemical compositions prior to the event is used as a reference value for

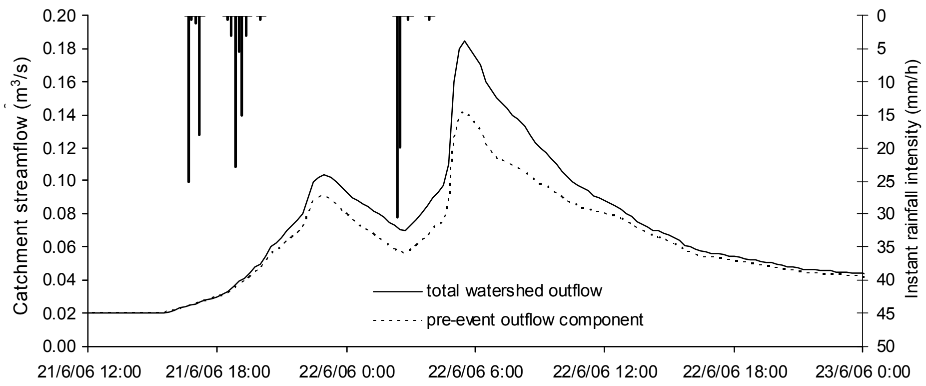

Figure 9. Outflow hydrograph at the stream gauging profile

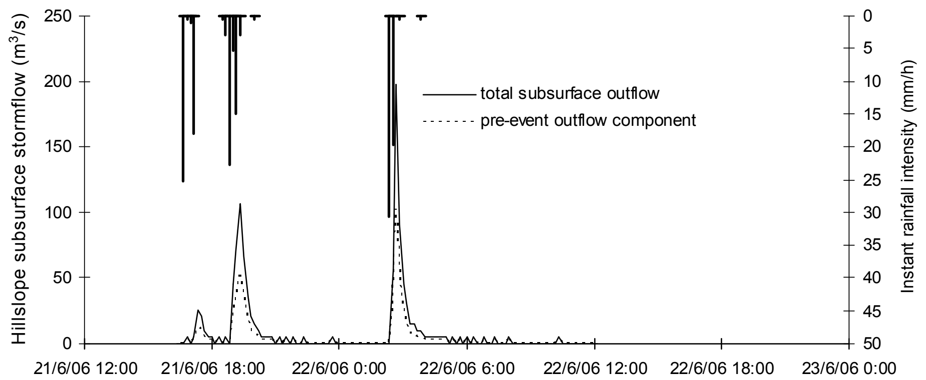

Figure 10. Outflow hydrograph at the subsurface trench

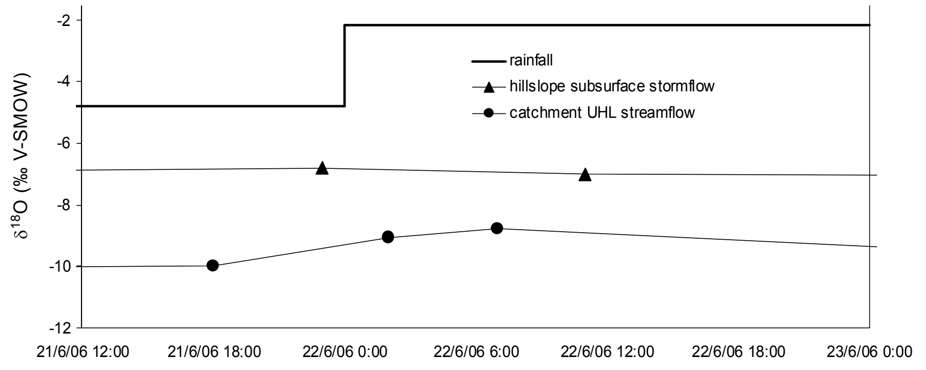

Figure 11. Isotope $\delta^{18}O$ in rain and subsurface and stream stormflow

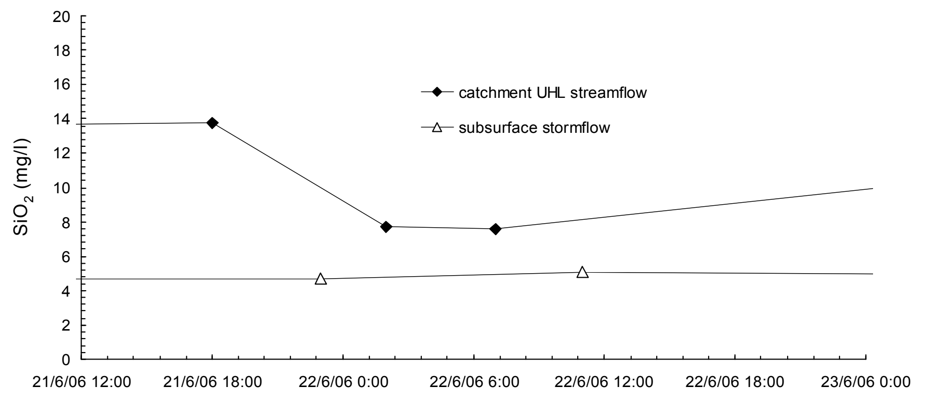

Figure 12. Silica in rain and subsurface and stream stormflow

the pre-event water in the rainfall-runoff episode evaluation. A strong buffering effect of the soil profile has been identified by means of isotopic and geochemical data. These findings have significantly expanded the knowledge of the hydrological role of shallow soils in the runoff formation and have better elucidated local water management questions on why no significant surface runoff occurres even after a radical deforestation. The approaches used in this study demonstrated the need for their conjunctive application, in particular when interpreting the origin and pathways of the quick runoff components to benefit the studies of water chemistry, fish species rehabilitation and changes of water cycle due to the global climate change. The results obtained are of importance for the water and landscape management of the temperate humid regions of Central Europe in view of the anticipated future changes in the natural climate and water regime.

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