



Organic matter dynamics and stable isotope signature as tracers of the sources of suspended sediment

Y. Schindler Wildhaber, R. Liechti, and C. Alewell

Institute for Environmental Geosciences, Basel, Switzerland

Correspondence to: Y. Schindler Wildhaber (yael.schindler@unibas.ch)

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Abstract. Suspended sediment (SS) and organic matter in rivers can harm brown trout *Salmo trutta* by affecting the health and fitness of free swimming fish and by causing siltation of the riverbed. The temporal and spatial dynamics of sediment, carbon (C), and nitrogen (N) during the brown trout spawning season in a small river of the Swiss Plateau were assessed and C isotopes as well as the C/N atomic ratio were used to distinguish autochthonous and allochthonous sources of organic matter in SS loads. The visual basic program *IsoSource* with $^{13}\text{C}_{\text{tot}}$ and ^{15}N as input isotopes was used to quantify the temporal and spatial sources of SS. Organic matter concentrations in the infiltrated and suspended sediment were highest during low flow periods with small sediment loads and lowest during high flow periods with high sediment loads. Peak values in nitrate and dissolved organic C were measured during high flow and high rainfall, probably due to leaching from pasture and arable land. The organic matter was of allochthonous sources as indicated by the C/N atomic ratio and $\delta^{13}\text{C}_{\text{org}}$. Organic matter in SS increased from up- to downstream due to an increase of pasture and arable land downstream of the river. The mean fraction of SS originating from upper watershed riverbed sediment decreased from up to downstream and increased during high flow at all measuring sites along the course of the river. During base flow conditions, the major sources of SS are pasture, forest and arable land. The latter increased during rainy and warmer winter periods, most likely because both triggered snow melt and thus erosion. The measured increase in DOC and nitrate concentrations during high flow support these modeling results. Enhanced soil erosion processes on pasture and arable land are expected with increasing heavy rain events and less snow during winter seasons due to climate change. Consequently, SS and organic matter in the

river will increase, which will possibly affect brown trout negatively.

1 Introduction

All streams carry some suspended sediment (SS) under natural conditions (Ryan, 1991). An increase of SS due to anthropogenic perturbation, however, has been observed in the last decades (e.g. Owens et al., 2005). Perturbation includes forestry, pasture and agricultural activities, which enhance soil erosion processes and hence the sediment delivery into rivers (e.g. Pimentel and Kounang, 1998). In addition, it is expected that the frequency and intensity of heavy rain events in middle Europe will increase due to climate change (IPCC, 2007), enhancing soil erosion triggered by water. According to model calculations, the sediment supply from the Alpine region into the Rhine basin, for example, is expected to increase by about 250 % (Asselman et al., 2003). Increasing SS loads in rivers generally leads to a higher fine sediment infiltration rate in the riverbed gravel (Greig et al., 2005; Zimmermann and Lapointe, 2005; Schindler Wildhaber et al., 2012). SS and fine sediment infiltration (SI) can provide a serious threat to aquatic ecosystems including phytoplankton, aquatic invertebrates, and salmonid fish (for a review see Bilotta and Brazier, 2008). Salmonid fish can be affected by SS in several ways. Their eggs develop in so-called redds, a shallow depression created by the female brown trout, where eggs and sperms are deposited. The female covers the fertilized eggs with gravel. While SS can directly impact health and fitness of free swimming fish (Newcombe and Jensen, 1996), fine sediment infiltration in the redds can induce siltation of the gravel resulting in a decrease

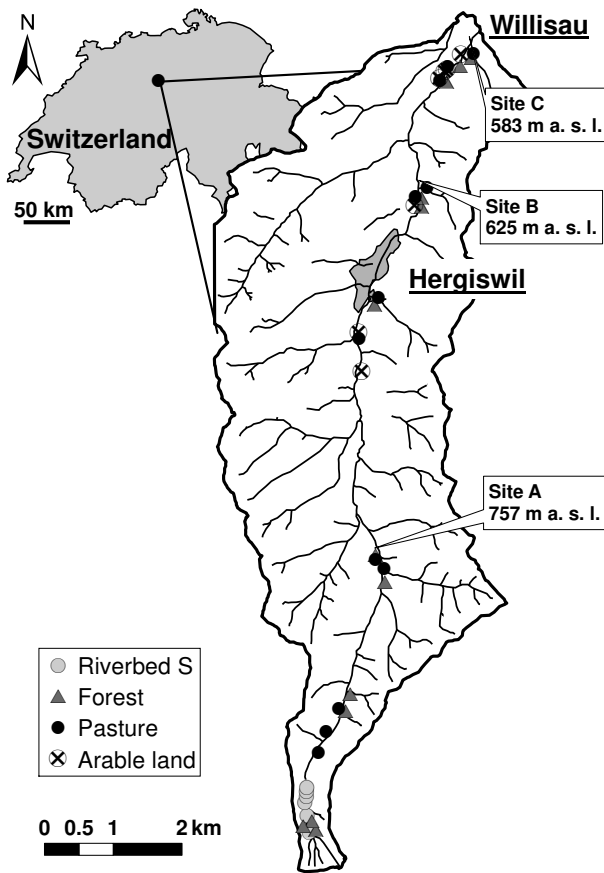


Fig. 1. Watershed of the river Enziwigger with the three field sites A, B and C including their altitude, soil sample spots and the towns Willisau and Hergiswil (Canton of Lucerne, Switzerland).

in hydraulic conductivity (Schälchli, 1995). This affects the oxygen supply to the developing salmonid embryos in the redds negatively, and hence their survival (Greig et al., 2005, 2007a; Heywood and Walling, 2007). The presence of high organic matter in the IS and interstitial water can additionally disproportionately impact on spawning habits (Greig et al., 2005). As respiration is strongly dependent on the availability of organic matter, oxygen demand within riverbeds will increase as the pool of organic matter increases (for a review see Greig et al., 2007b). Organic material is derived either from in-stream sources (autochthonous), for example macrophyte vegetation or from external sources (allochthonous), for example leaf litter or runoff from agricultural fields (Sear et al., 2008).

Schindler Wildhaber et al. (2012) reported in a study on sediment dynamics in a small Swiss headwater river of the Swiss Plateau with a native brown trout *Salmo trutta* population an SS increase from up- to downstream. This finding could be related to an increased shear stress attributable to a higher water level down the stream and/or to a higher fine sediment input from the arable land in the lower part of the catchment. Furthermore, organic carbon concentrations of

the SS were found to be highly variable with minimum values around 1.5 % at high flow and maximum values around 10.5 % at low flow (Schindler Wildhaber et al., 2012).

An identification of SS sources is required to improve site management and possibly restrain the described negative consequences for brown trout. Sediment tracer-based methods have been used to distinguish possible sources in watersheds. Stable carbon (C) and nitrogen (N) isotopes and as well as the carbon to nitrogen atomic ratio (C/N_a) have been found to be reliable tracers in recent studies (Onstad et al., 2000; McConnachie and Petticrew, 2006; Fox and Papanicolaou, 2007; Gao et al., 2007; Fox et al., 2010). Stable isotope compositions as well as C/N_a are affected by many factors including soil depth, vegetation, climate and cultural history (Kendall, 1998). By assessing the ratio of ^{13}C to ^{12}C , ^{15}N to ^{14}N and Ca to Na in the SS as well as in surface soils of the catchment, conclusions concerning possible SS origin can be drawn. The isotopic compositions of SS samples may represent a mixture of potential sources. The proportional contributions of different sources can be assessed by linear mixing models. The disadvantage of these models is generally the limitation in detecting potential sources by the number of isotope tracers. With n isotope tracers, $n + 1$ potential sources can be detected. The model is mathematically under-determined if the number of potential sources exceeds $n + 1$, resulting in an equation system with less equations than unknown variables and therefore no single solution is possible (Phillips and Gregg, 2001). Phillips and Gregg (2003) developed a visual basic program called *IsoSource* to assess potential source contributions if more than $n + 1$ sources are present. The program examines in small increment steps all possible combinations of each source contribution, resulting in several feasible solutions. This program was used in this study to quantify source contributions to SS during the brown trout spawning season.

The objective of this study was (I) to assess the temporal and spatial C and N dynamics during the brown trout spawning season in suspended and infiltrated sediments and river as well as interstitial water, (II) the use of C_{org} isotopes as well as C/N_a to distinguish autochthonous and allochthonous sources of the organic matter in the SS, and (III) the use of C_{tot} and N isotopes as tracers to quantify the sources of SS with respect to time and space.

2 Materials and methods

2.1 Study site and general setup

The river Enziwigger is a small canalized river located near Willisau, Canton of Lucerne, Switzerland with a total watershed area of about 31 km² (Fig. 1). The flow regime of the Enziwigger is not affected by hydro-power facilities and there is no waste water treatment plant located above Willisau. Like most rivers in the Swiss Plateau, its

morphology is strongly modified: Only 5 % is close to natural or natural, 21 % is little affected and 74 % is strongly affected or even artificial, including terraces that have been inserted to prevent deep channel erosion and scouring of the bed during flood events (classified with the Swiss modular stepwise procedure for ecomorphology after Hütte and Niederhauser, 1998; EBP-WSB-Agrofutura, 2005).

The bedrock of the watershed consists of Upper Freshwater Molasse. The soil types are mainly (stagnic) Cambisols and Leptosols (classified according to WRB (IUSS, 2006)). The mean annual temperature in Willisau is 8.5 °C with a mean annual rainfall of 1050 mm. Mean annual rainfall on the peak of the mountain Napf, where the river Enzizwigger originates, is 1700 mm per year (1961–2007; data from MeteoSwiss).

SS, IS and water samples were collected at three sites named A, B and C; from up- to downstream (Fig. 1) during the brown trout spawning season from November 2009 to end of March 2010. The sites had an altitude of 757, 625 and 583 m above sea level. For more information regarding the characteristics of the river and the three sites, see Schindler Wildhaber et al. (2012).

2.1.1 Sample collection

SS were sampled with 6 time-integrated SS samplers at each site following Phillips et al. (2000). The aperture of the SS samplers were about 60–70 mm above the riverbed. IS samples were collected in 6 sediment baskets per site. SS samplers and sediment baskets were both emptied in a weekly interval. The basket's sediment was sieved with a 4 mm sieve and the basket refilled with the remaining coarse sediment during each sampling event. Sediments <4 mm were taken to the laboratory for further analyses. The term infiltrated sediment (IS) refers to the total sediment <2 mm infiltrated during one week in the sediment basket and suspended sediment (SS) refers to the total amount of sediment caught during one week in the SS samplers. For a detailed description and discussion of the used methods for SS and IS see Schindler Wildhaber et al. (2012).

Interstitial water samples for dissolved organic carbon (DOC) and nitrate analyses were obtained in approximately two week intervals in 12 mini piezometers per site. The mini piezometers were 300 mm deep and perforated in the lower 160 mm. They were designed after Baxter et al. (2003) and are described in detail in Schindler Wildhaber et al. (2012). The interstitial water samples represent the water at the depth of the buried eggs in natural redds. Water samples were filtered in the field with 0.45 µm filters and laboratory analyses were conducted the following day. Soil samples for isotope and organic matter analyses were collected in forest, pasture and arable land with erosion evidences (Fig. 1). Soil profiles were determined at each sampling spot to ensure sampling of representative areas. In total, 40 topsoil samples of the watershed were analyzed. Additionally, an algae sample of each

site, six manure samples close to site B and C, and 5 riverbed sediment samples from the upper most accessible reach of the river were collected. No surface water ran at the upper most accessible reach of the river during dry periods, which allowed sediment sampling with a simple corer. It is assumed that these sediments represent mainly the original bedrock molasse. Downstream riverbed sediments were not sampled, as those samples would possibly represent a mixture of the other four sources. Source samples that are not independent would complicate the modeling results.

Water level at the three sites was measured every 15 s with pressure transmitter probes (STS Sensore Technik, Sirnach) and logged at 10 min intervals. Air temperature and precipitation was measured near site B at the town Hergiswil (Fig. 1) by a private company (KELAG Künzli Elektronik AG) in 5 min intervals on behalf of the Canton of Lucerne. Air temperature for site A and C was calculated from these data by assuming an increase of mean air temperature by 0.6 ° per 100 m in altitude (Leser et al., 2005).

2.1.2 Sample analyses

Soil and sediment samples were dried at 40 °C. Grain size distributions of the IS and SS were assessed with the standardized sieve techniques, grains with a diameter <32 µm with a sedigraph (Micrometrics 100, Coulter Electronics, Germany). Grain size fractions were classified according to the German soil taxonomy: Clay: <2 µm, silt: 2–63 µm and sand: 63 µm–2 mm (Sponagel et al., 2005). Organic and total carbon (C_{org} and C_{tot}) of the IS was measured by a Leco RC612 multiphase analyzer. C_{org} , C_{tot} , total nitrogen (TN) and C and N isotopes of SS and soil samples were measured with a continuous flow isotope ratio mass spectrometer (Thermo Finnigan, Germany) in line with a FLASH Elemental Analyzer 1112 (Thermo Finnigan, Italy) following standard processing techniques. For C_{org} analyses, inorganic carbon was eliminated with HCl vapor. Stable isotope ratios are reported as δ values per mil (‰) as: $\delta X = [(R_{sample}/R_{standard}) - 1] \times 1000$ where X is ^{13}C or ^{15}N and $R = ^{13}C/^{12}C$ or $^{15}N/^{14}N$. Standard reference materials are PDB limestone for C and air for N. The samples were corrected to internal standards EDTA ($\delta^{13}C = -30.3$ ‰, $\delta^{15}N = -1.1$ ‰) and AO-1, ammonium oxalate ($\delta^{13}C = -17.0$ ‰ and $\delta^{15}N = 32.7$ ‰). The precision reported for $\delta^{13}C$ and $\delta^{15}N$ analyzes was 0.1 ‰. DOC was measured with a TOC-Analyzer 5000A (Shimadzu Corporation), nitrate with an ion chromatograph with a Metrosep A Supp 5 column (Metrohm AG).

2.1.3 Data interpretation

Differences between two groups of data were tested with the Student's *t*-test. ANOVA was performed to determine significant differences between three or more groups. The relationship between a response variable and a possible predictor

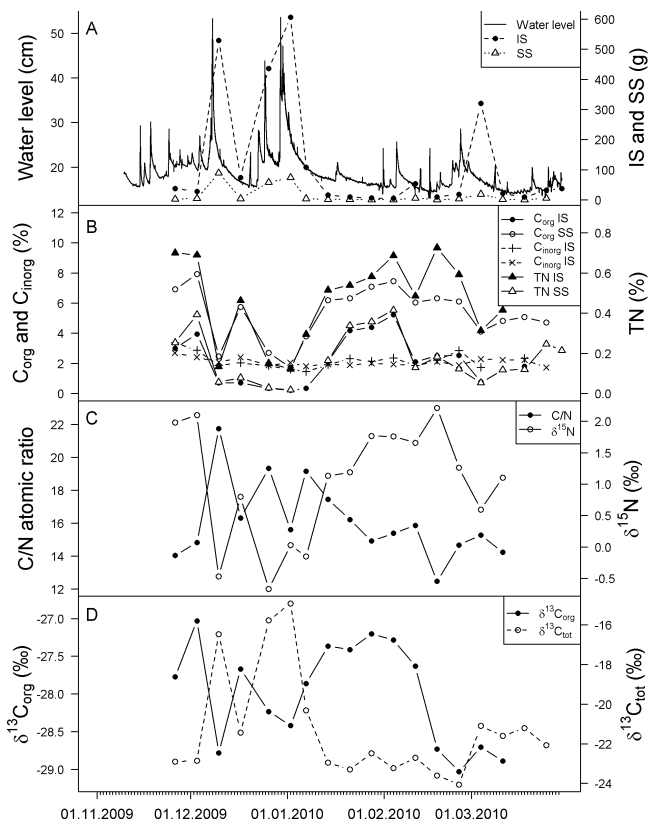


Fig. 2. Sediment, nutrient and isotope composition dynamics during the field period. Plotted are mean values of all samples from all three sites. **(A)** Water level at site B, the weekly infiltration of sediment <2 mm (IS) and the weekly suspended sediment (SS); **(B)** C_{org} and TN of IS and SS; **(C)** C/N and $\delta^{15}\text{N}$ of SS; **(D)** $\delta^{13}\text{C}$ of C_{org} and C_{tot} of the SS.

variable was assessed by linear regression models, backward stepwise linear regression models were used with several possible predictor variables. Relationships of data which were not normally distributed were tested with the Spearman rank correlation test. Significance level for all statistical analyses was set at 0.05.

The visual basic program *IsoSource* was used to quantify soil and sediment source contribution to SS (Phillips and Gregg, 2003). This program is available free at <http://www.epa.gov/wed/pages/models/stableIsotopes/isosource/isosource.htm> and has been successfully applied in different studies (e.g. Phillips et al., 2005; Gibbs, 2008; Phillips and Gregg, 2003). The program examines all possible combinations of each source contribution (0–100 %) in user-defined increment steps. Combinations that sum up to the measured isotopic compositions within a specified tolerance are considered to be feasible solutions from which the range of potential source contributions can be determined. Reporting the mean of the feasible solutions would only lead to misinterpretation of the results since every feasible solution may be the correct one (Phillips and

Gregg, 2003). Thus, the means of the possible solutions with standard deviations are reported in this study. Source increments of 1 % and mass balance tolerance levels of ± 0.1 ‰ were defined. In few cases, where no solution was found with these defaults, mass balance tolerance level was set to 1 ‰.

3 Results and discussion

3.1 Spatial and temporal dynamics of C_{org} in sediments and of DOC

C_{org} concentrations of the IS samples were significantly smallest at the upstream site A (mean = 1.7 ± 1.3 %). The highest mean C_{org} concentration in IS was found at site B (3.0 ± 2.5 %). IS at the most downstream site C had a mean C_{org} concentration of 2.3 ± 1.8 % (Table 1). Overall, the assessed C_{org} concentrations of the IS are relatively low. Sear et al. (2008) reported in a review organic matter concentrations of infiltrated fine sediment from 13 rivers in Europe. Concentrations ranged from 3.4 % to 24.5 % with a mean of 13 %. Heywood and Walling (2007) measured 15 % C_{org} in infiltrated fine sediment.

C_{org} concentration of the SS showed a similar spatial pattern as C_{org} concentration of the IS: the smallest concentrations were assessed at site A with a mean of 5.1 %, site B and C had significant higher concentrations with means of 6.6 and 6.5 %, respectively (Table 1). Again, these concentrations are relatively low. Acornley and Sear (1999) assessed 25 to 40 % C_{org} in the SS during low flows in summer and 15 to 20 % during high flows in autumn in two rivers in Hampshire, England, which were also brown trout spawning habitats. The relatively low C_{org} concentrations could partly be due to an instrumental bias of the SS samplers. Phillips et al. (2000) reported that SS samplers underestimate smallest SS particles to some extent. This would also include the light weighted organic material. They concluded, however, that the mean grain size does not statistically differ from point samples and that SS samplers collect statistically representative samples. The weekly assessed C_{org} concentrations of the SS are about four times higher than the concentrations in IS. This is probably due to the low specific gravity of organic material, holding it longer in suspension than inorganic material (Sear et al., 2008). The degradation of C_{org} is considered to be negligible (see Sect. 3.3). The increase of C_{org} in IS and SS from up to downstream can be explained by the higher percentage of agriculturally exploited land downstream of the watershed. Site A is surrounded by forest and pasture, while dominant land use at site B and C are arable farm land and pasture, which were both regularly manured.

C_{org} concentrations and the total amount of IS and SS showed remarkable inverse dynamics (Fig. 2). IS and SS generally increased with increasing water levels. C_{org} concentrations in IS and SS showed a significant inverse relationship

Table 1. Mean values and standard deviation at the three sites sampled of organic and inorganic carbon (C_{org} and C_{inorg}) in infiltrated sediment (IS) and suspended sediment (SS), dissolved organic carbon (DOC) in the river and the interstitial (int.), total nitrogen (TN) in IS and SS, nitrate in the river and interstitial, C/Na, $\delta^{13}\text{C}_{\text{org}}$, $\delta^{13}\text{C}_{\text{tot}}$ and $\delta^{15}\text{N}$ of SS. The sample numbers are given in parentheses.

	Site A	Site B	Site C
C_{org} IS (%)	1.7 ± 1.3 (29)*	3.0 ± 2.5 (30)*	2.3 ± 1.8 (27)
C_{inorg} IS (%)	2.1 ± 0.4 (29)	2.4 ± 0.8 (30)	2.1 ± 0.9 (27)
C_{org} SS (%)	5.1 ± 1.9 (29)	6.6 ± 2.7 (16)	6.5 ± 1.7 (27)
C_{inorg} SS (%)	2.1 ± 0.3 (29)	2.3 ± 0.7 (16)	2.0 ± 0.4 (27)
DOC river (mg l^{-1})	2.1 ± 0.6 (6)	1.8 ± 0.8 (7)	3.2 ± 1.7 (7)*
DOC int. (mg l^{-1})	2.0 ± 0.6 (36)*	2.5 ± 1.1 (27)	2.5 ± 1.0 (29)
TN IS (%)	0.2 ± 0.1 (17)	0.2 ± 0.2 (15)	0.2 ± 0.1 (14)
TN SS (%)	0.4 ± 0.1 (18)*	0.5 ± 0.3 (16)	0.5 ± 0.2 (12)
Nitrate river (mg l^{-1})	5.2 ± 0.7 (7)*	8.9 ± 0.6 (7)*	9.0 ± 1.2 (7)*
Nitrate int. (mg l^{-1})	4.9 ± 0.6 (34)*	8.9 ± 0.5 (27)*	9.0 ± 0.6 (29)*
C/N atomic IS	14.2 ± 2.0 (17)	16.8 ± 3.7 (15)	14.8 ± 2.9 (14)
C/N atomic SS	16.7 ± 2.1 (18)	15.3 ± 3.1 (16)	15.8 ± 2.1 (12)
$\delta^{13}\text{C}_{\text{org}}$ SS (‰)	-28.0 ± 0.5 (18)	-28.1 ± 0.9 (16)	-27.8 ± 0.9 (15)
$\delta^{13}\text{C}_{\text{tot}}$ SS (‰)	-20.1 ± 2.8 (18)*	-21.8 ± 2.6 (16)	-22.9 ± 3.0 (12)*
$\delta^{15}\text{N}$ SS (‰)	-0.4 ± 0.9 (18)*	2.0 ± 1.1 (16)*	1.8 ± 0.6 (12)

* Differs significantly from the two other sites (ANOVA, $p < 0.05$).

with water level and total IS and SS (Fig. 3a, b). Lowest C_{org} concentrations in IS (about 0.1 %) were measured during high flows in January and peak concentrations during base flows in February (about 6 to 8 %; Fig. 2a). Minimum C_{org} concentrations of the SS were also measured in January and were around 1.5 % and maximum at the end of February with concentrations around 10 % (Figs. 2b and 3b). A decrease of particulate organic carbon with increasing SS is reported in studies worldwide (Onstad et al., 2000; Meybeck, 1982; Gao et al., 2007; Zhang et al., 2009). The pattern can be explained by a dilution of C_{org} during high sediment loads with mineral matter derived from terrigenous soil erosion or remobilizing of mineral matter of the riverbed (Zhang et al., 2009). C_{org} has a very low specific gravity holding it longer in suspension than inorganic material, thus during base flow, the C_{org} proportion increases compared to heavier inorganic material. The significant relationship between the concentration of C_{org} in IS and SS and the silt and clay fraction (sediment $< 63 \mu\text{m}$) of IS and SS support this assumption (Fig. 3c). Silt and clay are also held in suspension more easily than sediment in the sand fraction.

It is assumed that the concentrations of C_{org} in the IS and SS are low enough that they do not have negative effects on the developing brown trout embryos in the river Enziwigger. Only during long periods of low flow with enhanced C_{org} concentrations in the SS and IS, C_{org} might induce a negative environments for the brown trout embryos. A high concentration of C_{org} can additionally block interstitial pore spaces, reduce gravel permeability and promote the growth of biofilms, leading to a decrease of oxygen availability (Greig et al., 2005).

The mean values of the measured DOC concentrations of the river and interstitial water samples were around 2 mg l^{-1} at all sites (Table 1). No significant differences in DOC concentrations in the river water and the interstitial water close to the brown trout eggs were assessed. DOC values around 2 mg l^{-1} represent a good mean water quality during the measured winter period in terms of DOC classified with the Swiss modular stepwise procedure for chemistry (Liechti, 2010). DOC values of the measured samples at site A never exceeded 4 mg l^{-1} ; thus, they were always in the category “good water quality” (Liechti, 2010). DOC values in river and interstitial water at site B and C exceeded 4 mg l^{-1} at three out of seven sampling dates. This indicates a “moderate” water quality in terms of DOC (Liechti, 2010). One of these relatively high values was assessed at the beginning of December during a high flow following strong precipitation. During heavy precipitation, previously unsaturated layers of the soil and river channel get connected to the drainage network. This delivers further carbon to the stream as new sources of DOC in upper organic soil horizons are assessed with rising water tables (Dawson et al., 2010). Otherwise, DOC can also be exported with near-surface soil runoff and/or overland flow generated during the storm event (Inamdar et al., 2004). DOC values $> 4 \text{ mg l}^{-1}$ were also measured in February during low flow conditions but with relatively high temperature (mean daily temperature around 5°C), which followed a cold period with mean temperature below freezing point. Biological activity might have been increased due to the higher temperature, resulting in an increase of DOC (Dawson et al., 2008) or DOC is flushed out from arable land to the river through infiltrating melt water (Hornberger et al., 1994).

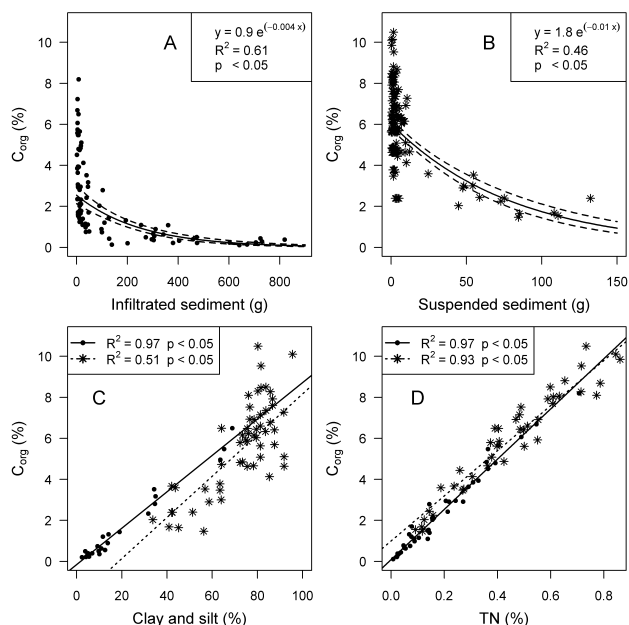


Fig. 3. Relationship between C_{org} and (A) total IS <2 mm; (B) total SS; (C) clay and silt fraction of sediment <2 mm and (D) TN. Solid circles and solid lines: Infiltrated sediment, stars and dotted lines: SS. Dashed lines in (A) and (B) are the 95 % confidence intervals.

3.2 Spatial and temporal dynamics of TN in sediments and of nitrate

TN increased similar to C_{org} from the upstream site A to the two downstream sites B and C (Table 1), most probably due to the increased sediment input from manured arable land and pasture. The increase of TN in the SS from site A with a mean of 0.4 % to site B and C with means around 0.5 % was significant (Table 1). The percentage of TN in SS was about 4 times higher than in the IS, thus showing the same pattern as C_{org} . The temporal dynamic of TN concentrations of IS and SS showed the same characteristics as the C_{org} dynamic (Fig. 2b): low levels at high discharge and high sediment yield and high levels at low discharge and low sediment yields (see previous section for explanation of this pattern). Overall, a highly significant relationship between TN and C_{org} concentration of captured sediments was found (Fig. 3d). The linear regressions between C_{org} and TN gives a y-intercept for IS of $0.09 (\pm 0.08)$ and for SS of 1.0 ± 0.2 (Fig. 3d). These small intercepts indicate mainly organic TN in these samples (Onstad et al., 2000).

Nitrate concentrations in the river and the interstitial water increased significantly from site A with a mean of 5.2 and 4.9 mg l^{-1} , respectively, to site B and C with means for river and interstitial water of 8.9 and 9.0 mg l^{-1} (Table 1). Nitrate concentrations for the two arable sites B and C did not differ significantly. A nitrate concentration below 5.6 mg l^{-1} represents a “good” water quality with respect to nitrate accord-

ing to the Swiss modular stepwise procedure for chemistry (Liechti, 2010). All but one measurement at site A was below this threshold concentration. The only sample exceeding this concentration was taken at high flow. This indicates nitrate leaching during high flow from the nearby pasture. Nitrogen leaching during storm events was described by other authors (e.g. Inamdar et al., 2004; Wagner et al., 2008). All samples at site B and C exceeded the concentration of 5.6 mg l^{-1} and two and three out of seven samples, respectively, even exceeded 8.4 mg l^{-1} . 8.4 mg l^{-1} is set as threshold concentration between the category “moderate” and “unsatisfying” water quality in terms of nitrate (Liechti, 2010). Thus, the nitrate concentrations of the river and interstitial water are too high, which is most probably due to manuring of the arable land and pasture at the two downstream sites. Nitrate concentrations in the interstitial water at the depth of brown trout eggs were not significantly different from nitrate concentrations in the river water. The lack of chemical gradients might indicate strong river water flux to the brown trout eggs (Malcolm et al., 2003).

3.3 Spatial and temporal dynamic of C/Na

Mean C/Na of SS are between 12.5 and 21.8 (Fig. 2c). C/Na generally decrease with increasing decomposition while $\delta^{15}N$ values increase (Conen et al., 2008). The correlation between mean C/Na and mean $\delta^{15}N$ of SS is significantly negative ($\rho = -0.8$). This inverse relationship can also be seen in Fig. 2c.

C/Na of the IS do not differ significantly from C/Na of the SS. Mean C/Na were between 14.2 and 16.8 for IS and SS at all three sites (Table 1). Values are similar to data reported for worldwide rivers (Meybeck, 1982), continental US rivers (Onstad et al., 2000), and in the Zhujiang River, China (Zhang et al., 2009). Mean C/Na of arable land, pasture and forest soil samples were between 10.7 and 16.8 (Table 2). C/Na of the analyzed algae as autochthonous sources of organic matter were significantly smaller with a mean of 9.1 ± 0.8 . The highest concentration of autochthonous organic matter would most likely occur during low flow conditions. No significant correlations between C/Na and SS or IS and between C/Na and water level at the sites were found. Consequently, the measured C/Na of IS and SS pointed to an allochthonous origin (e.g. soil or litter) of the organic matter in sediment. C/Na were excluded from further sediment tracer modeling because of the missing significant relationships between C/Na of the SS and water levels at the sites.

3.4 Spatial and temporal dynamics of C_{inorg} in sediments

IS and SS had a C_{inorg} concentration around 2 % at all sites (Table 1, Fig. 2b) due to carbonaceous bedrocks in the area. No significant differences between C_{inorg} concentrations of the IS and SS as well as between the three sites were found,

Table 2. Range (mean) of C/Na, $\delta^{13}\text{C}_{\text{org}}$, $\delta^{13}\text{C}_{\text{tot}}$ and $\delta^{15}\text{N}$ values of algae, manure, and riverbed sediment (riverbed S) as well as of forest, pasture and arable land soils of the watershed.

	C/Na	$\delta^{13}\text{C}_{\text{org}}$ (‰)	$\delta^{13}\text{C}_{\text{tot}}$ (‰)	$\delta^{15}\text{N}$ (‰)
Algae ($n = 4$)	7.9–9.6 (9.1)*	–41.4––31.1 (–35.0)*	–40.1–16.7 (–24.3)	–0.7–3.5 (2.1)*
Manure ($n = 6$)	14.6–34.9 (20.8)*	–28.7––25.3 (–27.9)	–29.0––25.7 (–27.9)	7.4–11.9 (8.9)*
Riverbed S ($n = 5$)	12.1–25.5 (17.8)	–28.1––26.7 (–27.3)	–3.2––0.7 (–1.9)*	–5.9––3.44 (–4.7)*
Forest ($n = 14$)	13.4–31.8 (16.8)	–28.4––26.8 (–27.5)*	–28.7––11.0 (–23.0)	–4.0–4.7 (–0.8)*
Pasture ($n = 12$)	11.4–26.1 (13.9)	–29.2––27.8 (–28.6)	–29.5––20.7 (–27.6)*	–1.0–6.4 (3.0)
Arable land ($n = 8$)	9.5–11.3 (10.7)*	–28.5––27.1 (–27.8)	–28.3––19.9 (–25.9)	4.3–7.7 (6.0)*

* Differs significantly from the five other potential sources (ANOVA, $p < 0.05$)

which is consistent with a steady pH value of the river water around 8.2 at all site. The C_{inorg} concentrations varied only marginally during the season and no correlation between C_{inorg} concentrations and the total IS, SS and the maximal water level during the week was assessed (Fig. 2b). Consequently, the concentration of C_{inorg} of IS and SS can not be used to draw any conclusions about the origin of the sediments.

3.5 Carbon and nitrogen isotopes for tracing suspended sediment sources

$\delta^{13}\text{C}_{\text{org}}$ values of SS were around -28.0 ± 0.9 ‰ and did not differ significantly between the three sites (Table 1). $\delta^{13}\text{C}_{\text{org}}$ values of soil and sediment samples from the catchment were in the same range (Table 2). Algae were highly depleted in $^{13}\text{C}_{\text{org}}$ resulting in $\delta^{13}\text{C}_{\text{org}}$ values between –41 and –31 ‰. Thus, $^{13}\text{C}_{\text{org}}$ isotopes indicate an allochthonous origin of the organic matter in SS, supporting the conclusion drawn from C/Na. $\delta^{13}\text{C}_{\text{tot}}$ values of the SS decreased significantly from upstream (mean site A: –20.0 ‰) to downstream (mean site C: –22.9 ‰), indicating different source contributions to the sediments (Table 1). The significantly higher $\delta^{15}\text{N}$ values at sites B and C with means of 2.0 ± 1.1 and 1.8 ± 0.6 ‰, respectively, support this assumption ($\delta^{15}\text{N}$ mean of site A = -0.4 ± 0.9 ‰, Table 1).

SS isotope compositions varied highly during the brown trout spawning season. $\delta^{13}\text{C}_{\text{tot}}$ values increased with higher water level and higher amount of SS while $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ values decreased (Fig. 2c, d). This indicates different SS sources related to the discharge pattern. Linear regression models showed significant relationships for all three sites between water level and $\delta^{13}\text{C}_{\text{tot}}$ values ($R^2 = 0.51$ to 0.95) and between water level and $\delta^{15}\text{N}$ values ($R^2 = 0.51$ to 0.66 , Fig. 4). No significant relationship was found between water level and $\delta^{13}\text{C}_{\text{org}}$. The relatively small range of $\delta^{13}\text{C}_{\text{org}}$ values (–29.9 to –26.5 ‰) compared to $\delta^{13}\text{C}_{\text{tot}}$ (–25.6 to –13.8 ‰) and $\delta^{15}\text{N}$ (–2.2 to 3.6 ‰) might be a reason for this missing significance. For this reason only, C_{tot} and N isotope compositions were used for further tracer modeling and $\delta^{13}\text{C}_{\text{org}}$ values were excluded.

Riverbed sediment in the upper watershed had $\delta^{13}\text{C}_{\text{tot}}$ values as high as –0.7 ‰ due to enrichment with carbonate. The carbonate contents of the sediment were between 2.2 and 3.4 %. In parallel, ^{15}N was depleted resulting in $\delta^{15}\text{N}$ values around –5 ‰ (Table 2, Fig. 5). These low values indicate young, poorly-decomposed material (Conen et al., 2008). The riverbed sediment probably originates mainly from the bedrock molasse. Very low C_{org} concentrations with a mean of 0.2 % support this assumption. $\delta^{13}\text{C}_{\text{tot}}$ of the forest soils varied highly (Table 2). Two of the six forest samples above site A had carbonate contents around 1.5 %. Those two soils were enriched in $^{13}\text{C}_{\text{tot}}$ resulting in $\delta^{13}\text{C}_{\text{tot}}$ values around –14 ‰. The remaining forest soil samples contained no carbonate resulting in $\delta^{13}\text{C}_{\text{tot}}$ similar to $\delta^{13}\text{C}_{\text{org}}$ around –28 ‰ (Fig. 5). $^{13}\text{C}_{\text{tot}}$ values of the forest soils upstream of site B and C were around –26 ‰. Again, carbonated soils (C_{inorg} around 0.3 %) were enriched in $^{13}\text{C}_{\text{tot}}$ compared to decarbonated soils. Mean $\delta^{15}\text{N}$ of the forest soil samples above site A was –2.5 ‰. The downstream forest soil samples around site B and C were enriched with ^{15}N (mean $\delta^{15}\text{N} = 1.5$ ‰). A decrease of $\delta^{15}\text{N}$ with increasing elevation and declining temperature has been assessed in several studies (for a review see Amundson et al., 2003) and can be explained by poorly-decomposed material. The steep slopes of the forests above site A can have an additional influence on the low $\delta^{15}\text{N}$ values. The soil residence time decreases with increasing slope and therefore $\delta^{15}\text{N}$ values decrease (Amundson et al., 2003).

$\delta^{13}\text{C}_{\text{tot}}$ values of pasture and arable land soil samples were between –19.9 and –29.5 ‰ at all sites (Table 2, Fig. 5). Differences between sites and between pasture and arable land in $\delta^{13}\text{C}_{\text{tot}}$ were not significant. Again, higher $\delta^{13}\text{C}_{\text{tot}}$ values were assessed in carbonaceous soils. $\delta^{15}\text{N}$ of pasture and arable land was significantly higher than in the forest. The significantly highest $\delta^{15}\text{N}$ values were assessed in arable land with a mean of 6.0 ‰ (Table 2). These high values are attributable to an acceleration of soil N loss through enhanced decomposition rates because of cultivation (Amundson et al., 2003). Additionally, manure is commonly enriched in ^{15}N (Amundson et al., 2003; Alewell et al., 2008). The manure samples, which were collected on arable land at site B and C, had a mean $\delta^{15}\text{N}$ of 8.9 ‰ (Table 2).

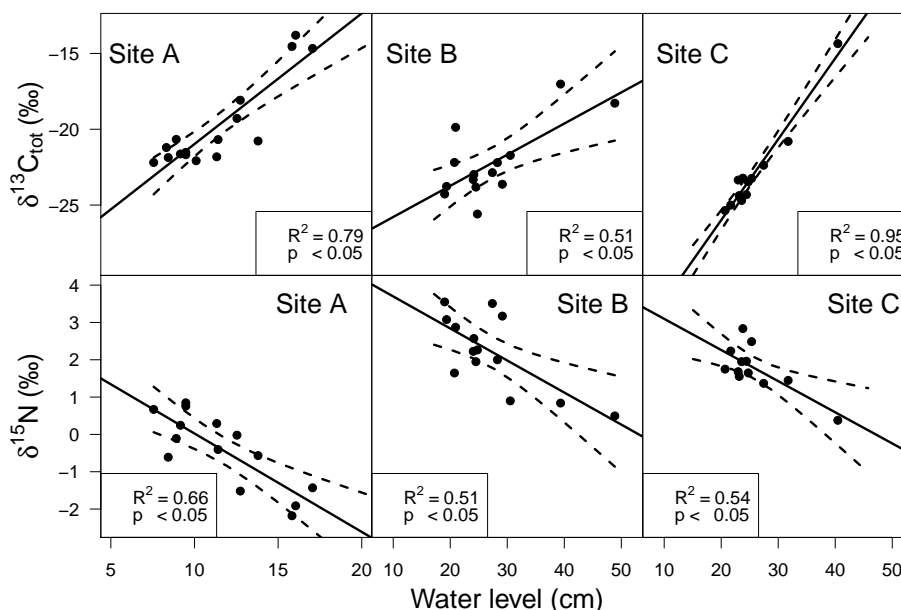


Fig. 4. Relationship between maximal mean daily water level of a week and $\delta^{13}\text{C}_{\text{tot}}$ and $\delta^{15}\text{N}$ values of the weekly captured SS at the three sites. Dashed lines are the 95 % confidence intervals.

These findings lead to the assumption that during high flow and high SS loads with high $\delta^{13}\text{C}_{\text{tot}}$ and low $\delta^{15}\text{N}$ values (Fig. 4), SS source is mainly the riverbed sediment in the upper watershed. During low flow and low SS loads with lower $\delta^{13}\text{C}_{\text{tot}}$ and higher $\delta^{15}\text{N}$ values (Fig. 4), SS sources are soils of forests, pasture and arable land (Fig. 5). The *IsoSource* program was used to quantify the proportion of the different sources (Phillips and Gregg, 2003). While interpreting the results, it is recommended to concentrate on the distribution of the feasible solutions rather than focus on a single value such as the mean to avoid misrepresenting the uniqueness of the results (Phillips and Gregg, 2003).

Algae samples were excluded from the modeling because C/Na and $\delta^{13}\text{C}_{\text{org}}$ values indicated none or minimal autochthonous C contributions. As described in Sect. 2.1.1, riverbed sediment samples were only taken in the upper most accessible reach of the river. Fox et al. (2010) noted that transformation of nitrogen could occur during temporary storage of sediments in the streambed, thus potentially masking their provenance. The sediment study in the Enzizwigger was only conducted during winter time when biological activity is low. Consequently, it is assumed that N and C_{tot} isotopic compositions would not significantly change during the temporary storage in the river. With biotic fractionation, higher isotope values and lower C/Na values with less organic concentrations are expected. The opposite was measured: With less organic material in the sediments, $\delta^{15}\text{N}$ values decreased and C/Na increased (Fig. 2). Additionally, decomposition processes triggered higher isotope values with smaller grain sizes since larger soil particles break into smaller particle sizes during these decomposition processes

(e.g. Fox and Papanicolaou, 2007). This was not the case for $\delta^{13}\text{C}_{\text{tot}}$, but it did apply to $\delta^{15}\text{N}$. This is, however, probably not attributable to physical fractionation but due to the fact that the mean sediment size as well as the source contributions varied in dependence of the water level.

SS at site A can possibly originate from forest and pasture soils or from riverbed sediment of the upper watershed. $\delta^{13}\text{C}_{\text{tot}}$ and $\delta^{15}\text{N}$ values of the three possible sources are clearly distinguishable (Fig. 5), resulting in well-defined *IsoSource* outcomes (Fig. 6). During base flow, the majority (up to $75 \pm 1\%$) of the SS originated from pasture soils. These percentages were relatively constant during the entire spawning season with an overall mean of 57 %, even though the catchment was temporarily covered with snow (Fig. 6, Table 3). The percentage of SS deriving from the upper watershed riverbed sediment increased significantly with increasing maximal mean daily water level of a week (linear regression, $R^2 = 0.69$). The highest values with up to $52.8 \pm 0.8\%$ of the SS originating from the riverbed sediment in the upper watershed were measured during the high discharge events in December and January with high SS loads. In general, the smallest part of the SS at site A originated from forest soils with an overall mean of 16 % (Table 3). The contribution of forest soils increased only after periods of higher temperature and thus snow melting periods (Fig. 5). Spearman rank correlation tests support this assumption with a significant correlation between SS deriving from forest and maximal mean daily temperature of a week ($\rho = 0.26$). These findings indicate the importance of forest-covered land to reduce SS delivery to the river to prevent negative effects of SS on the aquatic ecosystem.

Table 3. Mean (range and standard deviation) source contribution (%) to the suspended sediment at the three sites. Riverbed S = riverbed sediment.

Source	Site A	Site B	Site C
Riverbed S	27.0 (11.0 ± 0.7–52.8 ± 0.8)	18.3 (3.1 ± 2.0–41.2 ± 1.5)	12.1 (2.0 ± 1.5–48.2 ± 0.8)
Forest	16.0 (0 ± 1.7–45 ± 2.2)	16.8 (2.5 ± 1.9–49.1 ± 18.5)	26.4 (3.6 ± 3.1–40.9 ± 12.4)
Pasture	56.9 (28.4 ± 1.7–75 ± 1.0)	25.1 (3.8 ± 3.4–64.0 ± 14.6)	38.0 (6.7 ± 4.6–56.0 ± 20.7)
Arable land		39.9 (11.7 ± 5.1–70.4 ± 2.4)	23.5 (7.2 ± 4.5–45.8 ± 7.7)

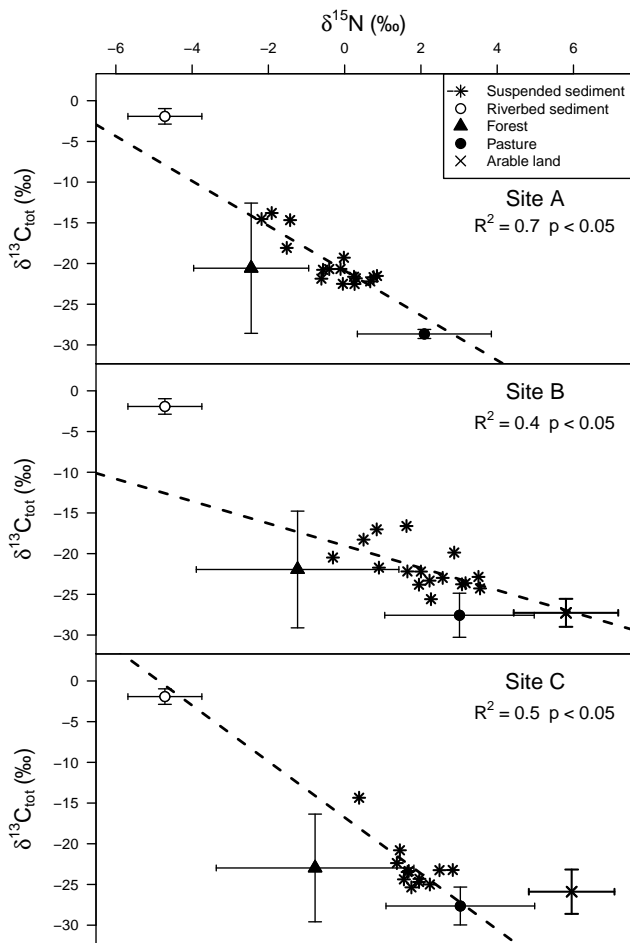


Fig. 5. $\delta^{13}\text{C}_{\text{tot}}$ and $\delta^{15}\text{N}$ values of SS and soil samples collected above each sites (average \pm sd). Dashed line: SS regression line.

Arable land represents an additional possible source for SS at sites B and C. The modeling results of these sites have higher uncertainty as there are four ($n + 2$) possible sources for the downstream sites with two (n) isotope groups and thus, an under-determined equation system. In addition, the distinction of the isotopic compositions of the possible sources was not as clear as at site A (Fig. 5). Nevertheless, some general conclusions were possible.

During base flow conditions, SS at site B originated mainly from arable land followed by pasture and forest soils

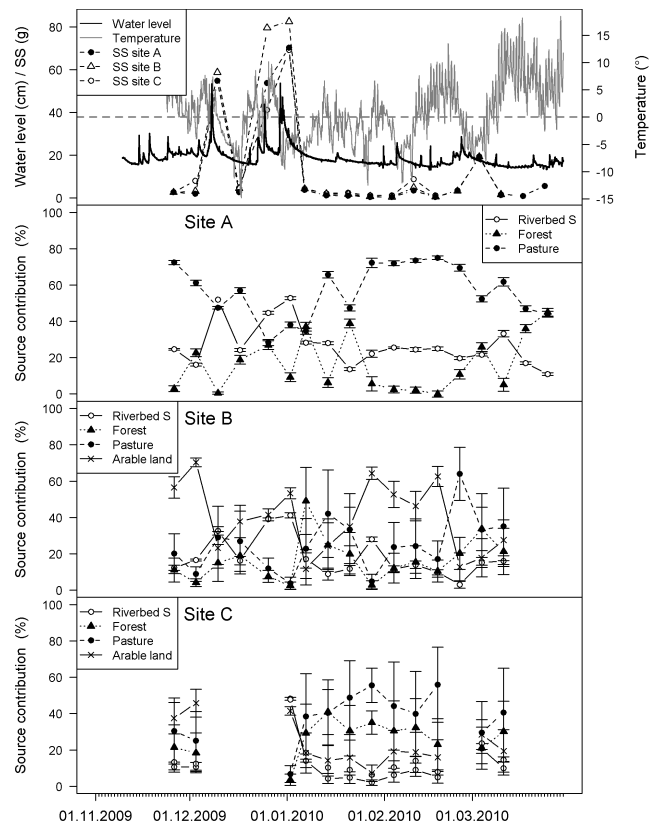


Fig. 6. Total suspended sediment (SS) per week at the three sites and soil source contribution from the three/four possible sources to SS at the three sites, determined with the dual isotope mixing model *IsoSource*. Air temperature was measured close to site B in Hergiswil.

(Fig. 6). On average, about 40 % of the SS originated from arable land (Table 3). The amount of SS originating from arable land increased significantly with increasing temperature ($\rho = 0.31$). The percentages were analyzed by multiple regression, using the highest mean daily temperature of a week (Temp), the maximal daily precipitation (Rain), and the quadratic terms of the two variables as regressors. The regression was a rather good fit ($R^2 = 0.56$) with an overall significant relationship and no interactions between the

variables ($F_{4,91} = 20.1$, $p < 0.05$):

$$\text{SS from arable land} = 29.7 + 4.5 \text{ Temp} - 0.35 \text{ Temp}^2 - 0.5 \text{ Rain} + 0.002 \text{ Rain}^2 \quad (1)$$

These results indicate erosion processes on the fallow fields during rainy periods when the fields were neither snow-covered nor frozen. During high flow conditions, riverbed sediment of the upper watershed was the main source of SS, resulting in a significant positive correlation between SS with riverbed sediment origin and water level ($\rho = 0.56$). The feasible contributions of forest and pasture soils to SS often overlapped, making the distinction between their contributions impossible. Consequently, the mean contribution values in Table 3 have to be regarded with caution. For this reason, statistics were conducted with the sum of the SS fraction originating from forest and from pasture soils. Spearman correlations indicate a significant decrease of SS originating from forest and pasture soils with increasing temperature ($\rho = -0.31$) and precipitation ($\rho = -0.21$). This increase of forest and pasture soils sediment contributions during drier and colder periods might indicate a transportation of SS with percolating water during snow-covered periods. Forest and pasture areas upstream to site B are mainly located on the hillside on the right side of the river where the bedrock is relatively close to the surface (Fig. 7). This probably triggers a relatively fast subsurface flow. Moist soil in the warmer season and ice formation during the winter on the right side of the channeled riverbed support the assumption that subsurface water and groundwater is draining from the hillside to the river. Arable land is located on the flat planes on the left side of the river. Groundwater modeling as well as observations of riverine groundwater (head, temperature and electric conductivity) indicate infiltration processes on the left-sided river board dominating the local groundwater flow regime (Huber et al., 2012) (Fig. 7). Thus, forest and pasture soils represent the main sediment sources at site B during undisturbed conditions. Thus, forest and pasture soils represent basically the main sediment sources at site B during undisturbed conditions. Sediments originating in arable land predominate, however, these natural processes due to erosion.

The majority of the SS during base flow conditions at site C came from pasture (29 ± 17 to 56 ± 20 %) and forest soils (21 ± 12 to 41 ± 12 %; Fig. 6). Both fractions increased significantly during colder and drier periods (Spearman rank correlations, pasture vs. precipitation $\rho = -0.39$, pasture vs. temperature $\rho = -0.37$, forest vs. precipitation $\rho = -0.41$ and forest vs. temperature $\rho = -0.61$.) Multiple regression models with maximal daily precipitation and mean daily temperature of a week could explain 58 % of the SS originating from pasture and 61 % of the SS originating from forest ($p < 0.05$). This indicates again a transportation of SS through percolating water from the hillside during snow-covered periods. The percentage of SS deriving from arable land was between 7.2 ± 4.5 and 45.8 ± 7.7 % with in-

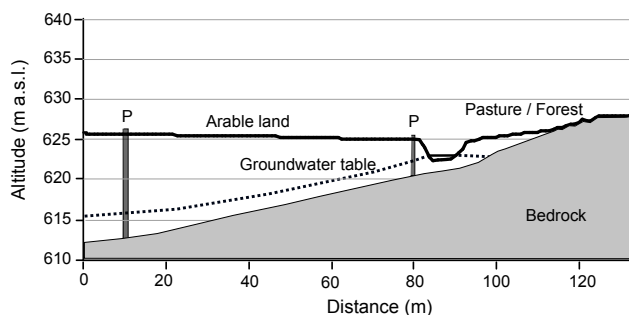


Fig. 7. Schematic view of the groundwater table and bedrock at site B with the installed piezometers (P).

creasing values with higher temperature (linear regression, $R^2 = 0.49$, $p < 0.05$) and precipitation (linear regression, $R^2 = 0.36$, $p < 0.05$; Table 3). Maximum daily precipitation and maximum daily mean temperature of a week together explain 74 % of the amount of SS originated from arable land (multiple regression, $p < 0.05$). Unfortunately, only one sample from an extreme flow event was assessed due to the loss of SS samplers. Nevertheless, the mean percentage of SS deriving from the upper watershed riverbed sediment depended significantly on the maximal daily mean water level (linear regression, $R^2 = 0.95$). During low flow conditions, riverbed sediment accounted for only 2 ± 1.5 % of the SS, during high flow for 48.2 ± 0.8 % (Table 3).

These observations have important implications for arable land management strategies. Anticipated warmer winters with more frequent heavy rain events (IPPC, 2007) are likely to enhance the SS delivery from arable land at site B and C as well as DOC and nitrate input to the river. Besides the negative effects on the arable land (loss of soil and organic material to the river), brown trout eggs can be negatively impacted by the enhanced input of SS and organic material (Greig et al., 2005, 2007b). A possible adaption could be a wider protecting strip next to the river with natural vegetation or greening of the arable fields during the winter.

4 Conclusions

The concentration of C_{org} as well as TN in the IS and SS varied highly during the brown trout spawning season. The highest values were assessed during low flow periods with small sediment loads, the lowest values during high flow periods with high sediment loads. This is suggested to be related to the dilution with mineral matter deriving from terrigenous soil erosion or the remobilizing of mineral matter of the riverbed. Organic matter concentrations of IS and SS are relatively low. Their impact on brown trout eggs is therefore expected to be low. The organic matter concentration of the sediment as well as nitrate in the river and interstitial water increased from the upstream site A to the two downstream sites B and C, which is attributable to leaching from

pasture and arable land. C/Na and $\delta^{13}\text{C}_{\text{org}}$ values indicate an allochthonous source of the organic matter in the SS during the brown trout spawning season. C_{tot} and N isotopes were used to trace the source of SS in respect to time and space using the visual basic program *IsoSource*. The fraction of SS originating from the riverbed sediment of the upper watershed increased at all sites during high flow. At site A, these fractions were the highest with values between $11.0 \pm 0.7\%$ to $52.8 \pm 0.8\%$ (mean = 27.0%). Smallest contributions from the riverbed sediment were detected at site C. The SS source contributions varied between sites during base flow conditions: at site A SS mainly originated from pasture, at site B mainly from arable land and pasture, and at site C from pasture and forest. Increasing winter temperatures and precipitation lead to a higher contribution of SS from arable land at both downstream sites, indicating soil erosion from the bare fields during snow-free and snow-melting periods. The increased DOC and nitrate concentration during high flow support these *IsoSource* calculations. These data indicate an increase of soil erosion processes on snow-free pasture and arable land during the anticipated warmer winter with more frequent torrential rain events (IPPC, 2007). An increase of SS and of organic matter concentration in the SS during the brown trout spawning season is a probable consequence. Both of these affect brown trout eggs negatively (Greig et al., 2005, 2007b).

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