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Organic matter dynamics and stable isotopes for tracing sources of suspended sediment

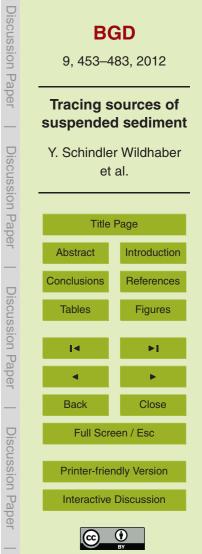
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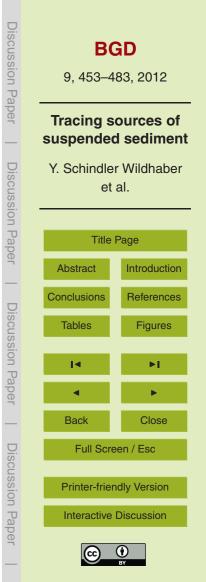
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Abstract

Suspended sediment (SS) and organic matter in rivers can harm brown trout *Salmo trutta* by impact on health and fitness of free swimming fish and siltation of the riverbed. The later results in a decrease of hydraulic conductivity and therefore smaller oxygen supply to the salmonid embryos. Additionally, oxygen demand within riverbeds will increase as the pool of organic matter increases. We assessed the temporal and spatial dynamics of sediment, carbon (C) and nitrogen (N) during the brown trout spawning season and used C isotopes as well as the C/N atomic ratio to distinguish autochthonous and allochthonous sources of organic matter in SS loads. The visual basic program *IsoSource* with ¹³C_{tot} and ¹⁵N as input isotopes was used to quantify the sources of SS in respect of time and space. Organic matter fractions 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 pre-

- ¹⁵ cipitation 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 $\delta^{13}C_{org}$. Organic matter in SS increased from up- to downstream due to pasture and arable land. The fraction of SS originating from upper watershed riverbed sediment increased at all sites during high flow. Its mean fraction decreased from up- to downstream. During
- ²⁰ base flow conditions, the major sources of SS are pasture and arable land. The later increased during rainy and warmer periods probably due to snow melting and erosion processes. These modeling results support the measured increased DOC and NO₃ concentrations during high flow.

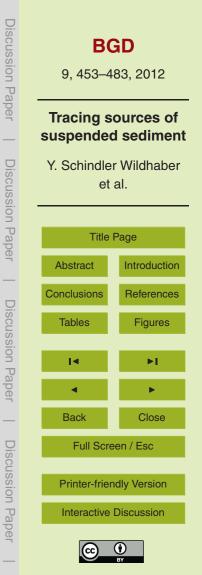


1 Introduction

All streams carry some suspended sediment (SS) under natural conditions (Ryan, 1991). However, an increase of SS due to anthropogenic perturbation 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 heavy rain events will increase due to climate change in middle Europe (IPPC, 2007), enhancing soil erosion triggered by water. According to model calculations, the sediment supply from the Alpine region in the Rhine basin for example is expected to increase by about 250 % (Asselman et al., 2003). Increasing SS loads in rivers generally lead to a higher fine sediment (sediment < 2 mm) deposition, possible resulting in riverbed clogging (Greig et al., 2005; Zimmermann and Lapointe, 2005). Fine sediment can provide a serious threat to aquatic ecosystems including phytoplankton, aquatic invertebrates and salmonid fish (for a review see Bilotta and Brazier.

- ¹⁵ 2008). SS can affect health and fitness of the free swimming fish (Newcombe and Jensen, 1996) and fine sediment deposition in the gravel bed can induce siltation of the riverbed resulting in a decrease of hydraulic conductivity (Schaelchli, 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). In ad-
- dition, the presence of high organic matter in the infiltrated sediment (IS) can have disproportionate impacts 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 from either in stream sources (autochthonous), for example
- ²⁵ macrophyte vegetation, or from external sources (allochthonous), for example leaf litter or runoff from agricultural practices (Sear et al., 2008).

Schindler Wildhaber et al. (2012) showed in a study on sediment dynamics in a small Swiss headwater river of the Swiss Plateau, which serves as recruitment sites for



brown trout *Salmo trutta*, an SS increase from up- to downstream. This finding could be related to an increasing shear stress due to higher water level down the stream and/or to a higher input of fine sediment from the arable land in the lower part of the catchment. Further, organic carbon fraction of the SS was 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 and organic matter sources is required to improve site management and possibly restrain the described negative consequences on brown trout. Sediment tracer-based methods with natural tracers have been used to distinguish possible sources in watersheds. Stable carbon and nitrogen isotopes ¹³C and ¹⁵N

as well as the carbon to nitrogen atomic ratio (C/N*a*) have been found to be reliably 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 values as well as C/N*a* are affected by many factors including soil depth, vegetation, climate and

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- ¹⁵ cultural history (Kendall, 1998). By measuring ¹³C, ¹⁵N and C/Na in the SS as well as in surface soils in the catchment, conclusions concerning possible SS origin can be drawn. The isotopic values of SS samples may represent a mixed value of different sources. The proportional contribution of different sources can be assessed by linear mixing models. The disadvantage of these models is generally the limitation in detect-
- ²⁰ ing possible sources by n + 1 sources with n different isotope groups. The model is mathematically underdetermined if the number of sources exceeds n + 1 resulting in a equation system with less equations than unknown variables and therefore no single solution is possible (Phillips and Gregg, 2001). This restriction has been overcome by Phillips and Gregg (2003) who developed a visual basic program called *IsoSource* which examines in small increment steps all possible combinations of each source
- which examines in small increment steps all possible combinations of each source contribution. We used this program 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 a headwater catchment, II) the use



of C_{org} isotopes as well as the C/N atomic ratio to distinguish autochthonous and allochthonous sources of the organic matter in the SS and III) the use of C and N isotopes as tracers to quantify the sources of SS in respect of time and space.

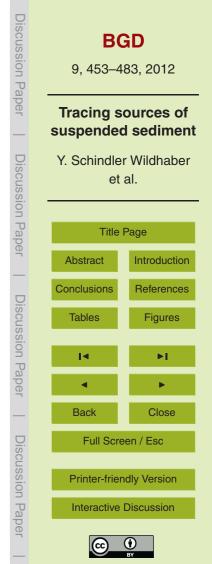
2 Materials and methods

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5 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 of about 31 km² (Fig. 1). The flow regime of the Enziwigger is not affected by hydro-power and no waste water treatment plant is located above Willisau. Like most rivers in the Swiss Plateau, its morphology is strongly modified though. Classified with the Swiss modular stepwise procedure for ecomorphology (Huette and Niederhauser, 1998), only 5 % is close to natural or natural, 21 % is little affected and 74 % is strongly affected or even artificial, including steps that have been inserted to prevent heavy bed scouring during flood events (EBP-WSB-Agrofutura, 2005).

- ¹⁵ The bedrock of the watershed consists of upper fresh water 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 mountain Napf, were the river Enziwigger originates, is 1700 mm per year (1961–2007; Data from MeteoSwiss).
- SS and IS 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 a.s.l. For more information regarding the characteristics of the river and the three sites see Schindler Wildhaber et al. (2012).



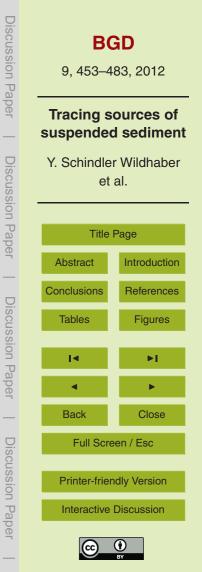
2.1.1 Sample collection

SS were sampled with 6 time-integrated SS samplers at each site following Phillips et al. (2000). 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 signed with a 4 mm signe and refilled with the same rest during each sampling.

- ⁵ was sieved with a 4 mm sieve and refilled with the coarse rest during each sampling event. Sediments < 4 mm were taken to the laboratory for further analysis. 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 (NO₃) analysis were obtained in approximately two week intervals in 12 mini piezometers per site. These mini piezometers were designed after Baxter et al. (2003) and are described in detail in Schindler Wildhaber et al. (2012). Water samples were filtered in the field with
- 0.45 µm filters and laboratory analysis 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 sample spot to ensure sampling of representative areas. In total, 40 topsoil samples of the watershed were analyzed. Additionally, 5 riverbed sediment samples from the upper most accessible reach of the river, an algae sample of each site and six manure samples
- close to site B and C were collected.

Water level at the three sites was measured every 15s with pressure transmitter probes (STS Sensore Technik Sirnach) and logged at 10 min intervals. Air temperature und precipitation was measured near site B at the town Hergiswil (Fig. 1) by a private

²⁵ company (KELAG Kuenzli Elektonik AG) in 5 min intervals on behalf of the Canton of Lucerne. Air temperature for site A and C was calculated from these data with the assumption that mean air temperature raises 0.6° per 100 m in altitude (Leser et al., 2005).



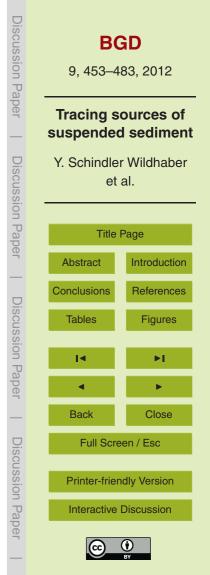
2.1.2 Sample analyses

Soil and sediment samples were dried at 40 °C. Grain size distribution of the IS and SS was assessed with the standardized sieve techniques, grains with a diameter $< 32 \,\mu$ m with a sedigraph (Micrometrics 100, Coulter Electronics, Germany). Grain s size fractions were classified according to the German soil taxonomy: Clay: $< 2 \mu m$, silt: 2-63 µm and sand: 63 µm-2 mm (Sponagel et al., 2005). Organic and total carbon (Corg and Ctot) of the IS was measured by a Leco RC612 multiphase analyzer. Corg, Ctot, 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) fol-10 lowing standard processing techniques. For Corg analyses, inorganic carbon was eliminated with HCl vapor. Stable isotope ratios are reported as δ values per mil (‰) as: $\delta X = [(R_{\text{sample}}/R_{\text{standard}}) - 1] \times 1000 \text{ where } X \text{ is } {}^{13}\text{C or } {}^{15}\text{N} \text{ and } R = {}^{13}\text{C}/{}^{12}\text{C or } {}^{15}\text{N}/{}^{14}\text{N}.$ Standard reference materials are PDB limestone for C and air for N. The samples were corrected to internal standards EDTA (δ^{13} C = 30.25 ‰, δ^{15} N = 1.1 ‰) and AO-1, 15 ammonium oxalate (δ^{13} C = -17.02 ‰ and δ^{15} N = 32.7 ‰). The precision reported for δ^{13} C and δ^{15} N analyzes was 0.1 ‰. DOC was measured with a TOC-Analyser 5000A (Shimadzu Corporation), NO₃ with an ion chromatograph with a Metrosep A Supp 5 column (Metrohm AG).

20 2.1.3 Data interpretation

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Differences between two groups of data were tested with the Student's *t*-test. Significance level for Student's *t*-tests and linear regression 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 freely availably at http://www.epa.gov/wed/pages/models/stableIsotopes/isosource/isosource.htm and was 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 to the observed isotopic signatures within a specified tolerance are considered to be feasible solutions from which the range of potential source contributions can be determined. Only reporting the mean of the feasible solutions would lead to misinterpretation
of the results since every feasible solution may be the correct one (Phillips and Gregg, 2003). We defined source increments of 1 % and mass balance tolerance levels of ±0.1 ‰. In few cases, where no solution was found with this defaults, mass balance tolerance levels was set to 1 ‰.

3 Results and discussion

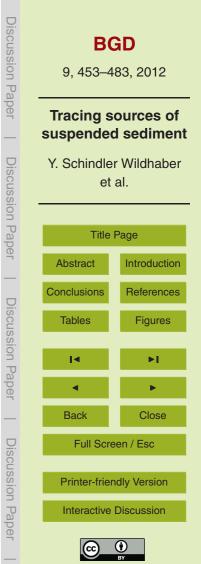
3.1 Spatial and temporal dynamics of C_{orq} in sediments and of DOC

The C_{org} fraction of the IS samples was smallest at the upstream site A (mean = 1.7 ± 1.3 %). The highest mean value was found at site B (3.0 ± 2.5 %). The most downstream site C had a mean value of 2.3 ± 1.8 % (Table 1). The differences between site B and C were not significant. Overall, the assessed C_{org} fractions of the IS are relatively low. Sear et al. (2008) reported in a review organic matter fractions of infiltrated fine sediment from 13 rivers in Europe. Values ranged from 3.4% to 24.5% with a mean of 13%. Heywood and Walling (2007) measured a C_{org} fraction in the infiltrated fine sediment of 15%.

C_{org} fraction of the SS showed the same spatial pattern as C_{org} of the IS: the smallest
 values were found at site A with a mean of 5.1 %, site B and C had significant higher
 values with means of 6.6 and 6.5 %, respectively (Table 1). Again, these values are
 relatively low. Acornley and Sear (1999) for example reported from two rivers in Hampshire, England, which were also brown trout spawning habitats, C_{org} fractions in the SS
 between 25 and 40 % during low flows in summer and 15 to 20 % during high flows in
 autumn. The weekly assessed C_{org} fraction of the SS is about four times higher than

²⁵ autumn. The weekly assessed C_{org} fraction of the SS is about four times higher than the fraction in IS. This is probably due to the low specific gravity of organic material,

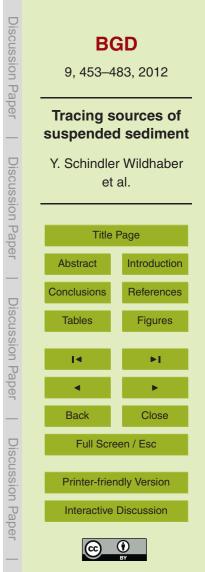
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holding it longer in suspension than inorganic material (Sear et al., 2008). The increase of C_{org} in IS and SS from up- to downstream can be explained by the higher percentage of the arable land downstream of the watershed. Site A is surrounded by forest and pasture, while dominant land use at site B and C are arable farm land and pasture, both of which were regularly manured.

Values of C_{org} , IS and SS showed remarkable parallel dynamics (Fig. 2): IS and SS generally increased with increasing water levels. C_{org} in IS and SS showed a significant inverse relationship with the water level and the total IS and SS (Fig. 3a,b). Lowest values of C_{org} in the IS (about 0.1%) were measured during high flows in January and peak values during base flows in February (about 6 to 8%; Fig. 2a). Minima of the C_{org} fractions of the SS were also measured in January and were around 1.5% and maxima end of February with fractions around 10% (Figs. 2b and 3b). A decrease of C_{org} and particulate organic carbon respectively 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

- ¹⁵ 2009). The pattern can be explained by a dilution of C_{org} during high sediment loads with mineral matter deriving 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. This can also be seen in the
- ²⁰ significant relationship between the fraction of C_{org} in IS and SS and the silt and clay fraction (sediment < 63 µm) of IS and SS (Fig. 3c). Silt and clay are also hold in suspension more easily than sediment in the size of sand. In addition, mechanical erosion with different intensity can reach different horizons of the soil profile. Low mechanical erosion mainly occurs at the surface soil horizon. With intensified soil erosion, soils
- in the deeper horizons with lower C_{org} can be transported into river systems. Thus, intensified erosion in the deeper soil horizons will result in lower contents of C_{org} in the river sediment (Zhang et al., 2009). Observed landslides in the catchment support this hypothesis.



The assessed DOC values of the river and interstitial water samples showed a small but not significant increase from the upstream site A to the two downstream sites. Mean values were around 2 mg I⁻¹ at all sites (Table 1). This represents 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 at site A never exceeded 4 mg I⁻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 I⁻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 beginning of December during a high flow following strong precipitation. This precipitation probably triggered surface erosion on fallow fields resulting in higher DOC in the river. A significant positive correlation between DOC and discharge has also been found in other studies in different catchments (e.g., Zhang et al., 2009; Dawson et al., 2010). DOC

values $> 4 \text{ mg l}^{-1}$ were also assessed 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 bellow freezing point. Due to the higher temperature bi-

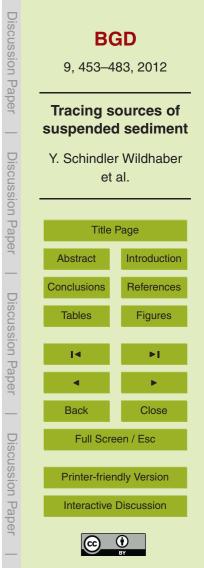
ological activity might rose, resulting in an increase of DOC (Dawson et al., 2008) or

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

DOC was transported through melting water from arable land to the river.

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- ²⁰ 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 increase of sediment input from manured arable land and pasture. The increase of TN in the IS is not significant (p = 0.06) due to a general small TN percentage of about 0.2%. 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 ²⁵ (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 the TN fraction of IS and SS showed also 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 the TN and the C_{org} fraction of captured sediments was found (Fig. 3d). The linear regression between C_{org} and TN gives a *y*-intercept for IS of 0.09 (±0.08) and for SS of 1.0±0.2 (Fig. 3d). These small intercepts indicate that most of the TN in these samples was organic (Onstad et al., 2000).

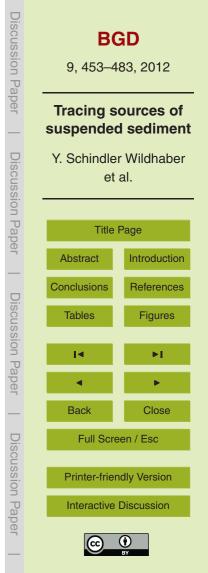
Nitrate in the river and the interstitial water increased significantly from site A with a mean of 5.2 and 4.9 mg I^{-1} respectively to site B and C with means for river and interstitial water of 8.9 and 9 mg I⁻¹ (Table 1). Nitrate values for the two arable sites B and C did not differ significantly. A nitrate content bellow 5.6 mg I⁻¹ represents a "good" water quality in respect of nitrate according to the Swiss modular stepwise procedure for chemistry (Liechti, 2010). All but one measurement at site A was bellow this threshold value. The only sample exceeding this value was taken at high flow. This indicates

- that nitrate was leached during high flow from the nearby pasture. All samples at site B and C exceeded the concentration of 5.6 mg l⁻¹ and two, respectively three out of seven samples even exceeded 8.4 mg l⁻¹. 8.4 mg l⁻¹ is set as threshold value 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 most probably due to manuring of the arable land and pasture at the two downstream sites.

20 3.3 Spatial and temporal dynamic of atomic C/N ratio

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The C/Na ratios of the IS did not differ significantly from the C/Na ratios of the SS. Mean C/Na were between 14.2 and 16.8 for IS and SS at all three sites (Table 1). This is similar to values reported for worldwide rivers (Meybeck, 1982), continental US rivers (Onstad et al., 2000) and in the Zhujiang River, China (Zhang et al., 2009). Mean
²⁵ C/Na ratios of arable land, pasture and forest soil samples were between 10.7 and 16.8 (Table 2). The C/Na ratios of the analyzed algae as autochthonous sources were significantly smaller with a mean of 9.1±0.8. The highest fraction of autochthonous organic matter would most likely occur during low flow conditions. We found neither



a significant correlation between C/Na and SS or IS nor with water level at the sites. Thus, the measured C/Na of IS and SS pointed to an allochthonous (e.g. soil or litter) origin of the organic matter in sediment. We excluded C/Na from further sediment tracer modeling due to the missing significant relationships between C/Na of the SS and water levels at the sites as well as between the sites.

Nevertheless, mean C/N*a* of SS varied between 12.5 and 21.8 (Fig. 2c). C/N ratios generally decrease with increasing decomposition while δ^{15} N values increase (Conen et al., 2008). We found a significant negative correlation ($R^2 = 0.66$) between mean C/N*a* and mean δ^{15} N of SS (Fig. 2c).

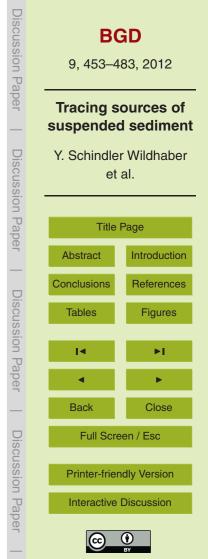
10 3.4 Spatial and temporal dynamics of C_{inorg} in sediments

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The fraction of C_{inorg} , which can be attributed to carbonaceous materials, in IS and SS was at all sites around 2% (Table 1, Fig. 2b). No significant differences between C_{inorg} in the IS and SS as well as between the three sites were found. The proportion varied only marginal during the season and no correlation between C_{inorg} and the total IS, SS and the maximal water level during the week was assessed (Fig. 2b). Thus, the fraction of C_{inorg} in IS and SS can not be used to draw any conclusions about the origin of the sediment.

3.5 Carbon and nitrogen isotopes for tracing suspended sediment sources

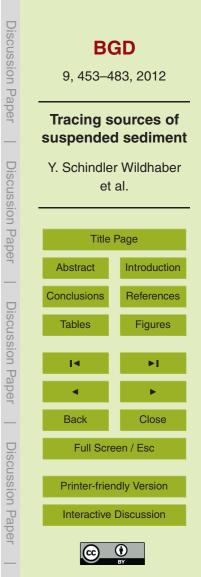
 $\delta^{13}C_{org}$ values of the SS did not differ significantly between the three sites. They were all around $-28.0 \pm 0.9 \%$ (Table 1) and thus in the same range as soil and sediment samples from the catchment (Table 2). Algae were highly depleted of $^{13}C_{org}$ resulting in $\delta^{13}C_{org}$ values between -41 and -31 %. Thus, isotopes indicate an allochthonous origin of the organic matter in SS, supporting the conclusion drawn from C/N*a* values. $\delta^{13}C_{tot}$ 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 δ^{15} N at sites B and C with means of



2.0 and 1.8%, respectively, support this assumption (δ^{15} N mean of site A = -0.4%, Table 1).

SS isotope values varied highly during the brown trout spawning season. $\delta^{13}C_{tot}$ increased with higher water level and higher amount of SS while $\delta^{13}C_{org}$ and $\delta^{15}N$ decreased (Fig. 2c,d). This indicates different SS sources in dependence on discharge. Linear regression models showed significant relationships for all three sites between water level and $\delta^{13}C_{tot}$ ($R^2 = 0.51$ to 0.95) as well as $\delta^{15}N$ ($R^2 = 0.51$ to 0.66, Fig. 4). No significant relationship was found between water level and $\delta^{13}C_{org}$. The relatively small range of $\delta^{13}C_{org}$ values (-29.9 to -26.5%) compared to $\delta^{13}C_{tot}$ (-25.6 to -13.8%) and $\delta^{15}N$ (-2.2 to 3.6%) might be a reason for this missing significance. For this reason we used $\delta^{13}C_{tot}$ and $\delta^{15}N$ for further tracer modeling and excluded $\delta^{13}C_{org}$. Consequently, we were only able to trace SS but not the organic matter in the SS.

Riverbed sediment in the upper watershed had $\delta^{13}C_{tot}$ values as heavy as -0.7%due to enrichment with carbonate. The carbonate contents of the sediment were be-15 tween 2.2 and 3.4 %. In parallel, there was a depletion of ¹⁵N with δ^{15} N values around -5‰ (Table 2, Fig. 5). These light values indicate young, poorly decomposed material (Conen et al., 2008). $\delta^{13}C_{tot}$ 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 ${}^{13}C_{tot}$ resulting in $\delta^{13}C_{tot}$ values around -14 ‰. The remaining 20 forest soil samples were decarbonated resulting in $\delta^{13}C_{tot}$ similar to $\delta^{13}C_{org}$ around -28% (Fig. 5). ¹³C_{tot} of the forest soils upstream of site B and C was around -26%. Again, carbonated soils (C_{inora} around 0.3%) were enriched of ¹³C_{tot} compared to decarbonated soils. The mean δ^{15} N value of the forest soil samples above site A was -2.5 %. The downstream forest soil samples around site B and C were enriched with ¹⁵N (mean $\delta^{15}N = 1.5$ %). A decrease of $\delta^{15}N$ 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 slope of the

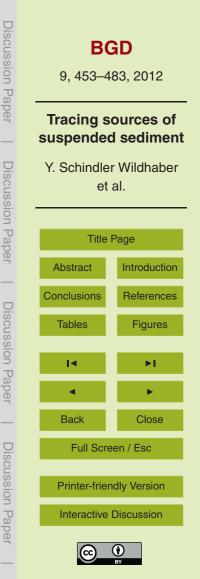


forests above site A can have an additional influence on the low δ^{15} N values. The soil residence time decreases with increasing slope and therefore δ^{15} N values decrease (Amundson et al., 2003).

- $\delta^{13}C_{tot}$ 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 $\delta^{13}C_{tot}$ were not significant. Again, higher $\delta^{13}C_{tot}$ values were assessed in carbonaceous soils. $\delta^{15}N$ of pasture and arable land was significantly higher than in the forest. The significantly highest $\delta^{15}N$ values were assessed in arable land with a mean of 6.0% (Table 2). This high value can be explained by an acceleration of soil N loss through enhanced decomposition rates because of cultivation (Amundson et al., 2003). Additionally, manure is commonly enriched in ¹⁵N (Amundson et al., 2003; Alewell et al., 2008). Our manure samples, which were collected on arable land, had a mean $\delta^{15}N$ of 8.9% (Table 2).
- These findings lead to the assumption that during high flow and high SS loads with high $\delta^{13}C_{tot}$ values and low $\delta^{15}N$ (Fig. 4), SS source is mainly the riverbed sediment in the upper watershed. During low flow and low SS loads with lower $\delta^{13}C_{tot}$ and higher $\delta^{15}N$ values (Fig. 4), SS sources are forests, pasture and arable land (Fig. 5). This assumption was tested with the *IsoSource* program (Phillips and Gregg, 2003). To interpret 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).

SS at site A can possibly originate from forest and pasture soils or from riverbed sediment of the upper watershed. $\delta^{13}C_{tot}$ and $\delta^{15}N$ values of the three possible sources are clearly distinguishable (Fig. 5), resulting in well-defined *IsoSource* outcomes (Fig. 6).

²⁵ During base flow, the majority (up to 75%) of the SS came 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). About 45 to 53% of SS originated in the riverbed sediment in the upper water-shed during the high discharge events in December and January with high SS loads.



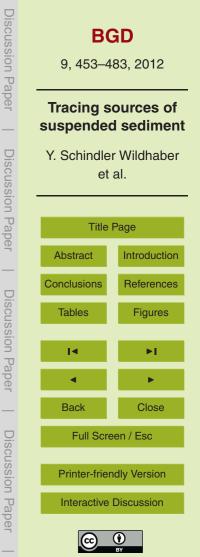
Linear regression analysis showed a significant correlation between maximum weekly water level and the percentage of SS deriving from the upper watershed riverbed sediment ($R^2 = 0.69$). In general, the smallest part of the SS at site A originated from forest soils with an overall mean of 16% (Table 3). The contribution of forest soils increased only after periods of higher temperature and thus snow melting periods (Fig. 5). Lin-

ear regression analysis support this assumption with a week but significant correlation between SS deriving from forest and weekly maximal temperature ($R^2 = 0.12$).

5

Arable land represents an additional possible source for SS at site B and C. Modeling results of these sites were connected with high standard deviations, making their interpretation more uncertain. The high standard deviations was due to the additional

- ¹⁰ interpretation more uncertain. The high standard deviation was due to the additional source, resulting in n+2 possible sources with n isotopes groups and thus an underdetermined equation system. In addition, the isotopic distinction between the possible sources was not as clear as at site A (Fig. 5). Nevertheless, some general conclusions were possible.
- ¹⁵ SS at site B originates mainly from arable land during base flow conditions, followed by pasture and forests (Fig. 6). On average, about 40% of the SS originates from arable land (Table 3). Multiple regressions showed a week but significant positive correlation of SS originating from arable land with higher temperature and precipitation ($R^2 = 0.12$), indicating erosion processes of the fallow fields during rainy periods when
- ²⁰ 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 and water level ($R^2 = 0.46$). The feasible contributions of forest and pasture soils to SS often overlap, making the distinction between their contributions impossible. Consequently the mean contribution values in Table 3 have
- ²⁵ to be regarded with caution. Multiple regression analyses show an increase of forest and pasture soils sediment contributions during drier and colder periods ($R^2 = 0.28$ for forest soils and 0.12 for pasture soils, p < 0.05). This might indicate a transportation of SS with percolating water during snow covered periods. Forest and pasture areas upstream to site B are to the majority 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. Icicles on the right side of the channeled riverbed support the assumption, that subsurface water and groundwater is exfiltrating from the hillside to the river. Arable land is located on the flat planes on the left side of the river.

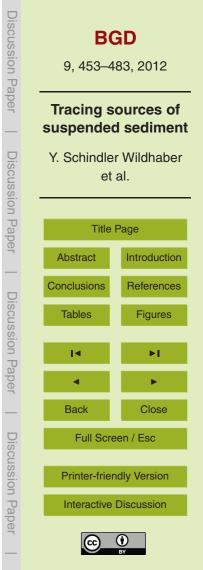
⁵ Groundwater modeling as well as observations of riverine groundwater (head, temperature and electric conductivity) indicate that on the left-sided river board infiltration processes dominate the local groundwater flow regime (Huber et al., 2012) (Fig. 7).

The majority of the SS during base flow conditions at site C came from pasture (29 to 56%) and forest soils (21 to 41%; Fig. 6). Both fractions increased significantly

- ¹⁰ during colder and drier periods (multiple regression: $R^2 = 0.58$ for pasture, $R^2 = 0.61$ for forest). This indicates again a transportation of SS through percolating water from the hillside during snow covered periods. Percentage of SS deriving from arable land were between 7.2 and 45.8% with increasing values with higher temperature ($R^2 = 0.49$) and precipitation ($R^2 = 0.36$, Table 3). Multiple regression analyses indicated, that ¹⁵ maximum precipitation and maximum weekly temperature explain together 74% of the
- amount of SS originated from arable land. Unfortunately, only one sample from an extreme flow event was assessed due to the loss of SS samplers. Nevertheless, we were able to see a significant dependency of the mean percentage of SS deriving from the upper watershed riverbed sediment on the water level ($R^2 = 0.95$). During low flow conditions, riverbed sediment accounted for only 2% of the SS, during high flow for 48% (Table 3).

4 Conclusions

The fraction of C_{org} as well as TN in the IS and SS varied highly during the brown trout spawning season. Highest values were assessed during low flow periods with small ²⁵ sediment loads, lowest values during high flow periods with high sediment loads. This is probably mainly due to dilution with mineral matter deriving from terrigenous soil erosion or remobilizing of mineral matter of the riverbed. Peak values in NO₃ and DOC



were measured during high flow probably due to water flow from pasture and arable land into the river. The organic matter fraction of the sediment as well as NO₃ in the river and interstitial water increased from the upstream site A to the two downstream sites B and C due to leaching from pasture and arable land. C/N atomic ratios and $\delta^{13}C_{ord}$ values indicate an allochthonous source of the organic matter in the SS dur-5 ing the brown trout spawning season. ¹³C_{tot} and ¹⁵N isotopes were used to trace the source of SS in respect of 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 10 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

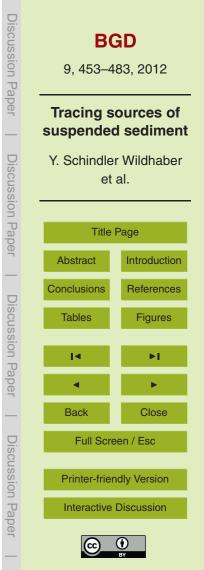
arable land at both downstream sites, indicating soil erosion from the bare fields during snow free and snow melting periods. The increased DOC and NO₃ concentration during high flow support these *IsoSource* calculations.

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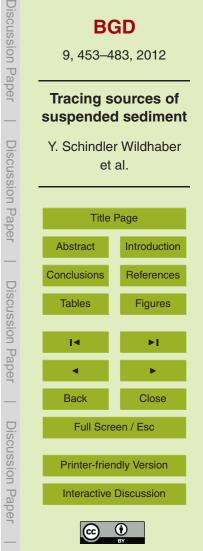
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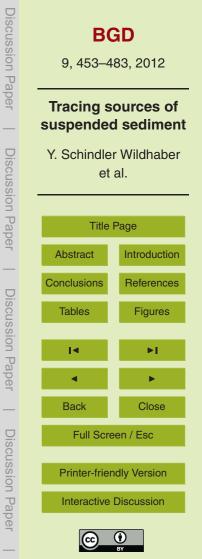
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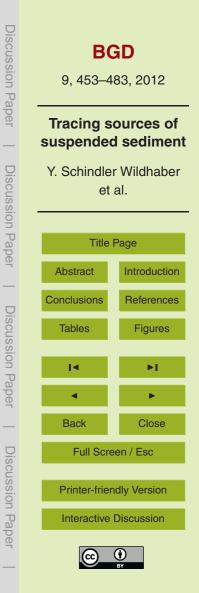
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Table 1. Mean ± standard deviation at the three sites of organic and inorganic carbon (C_{org} and C_{inorg}) in infiltrated sediment (IS) and suspended sediment (SS), dissolved organic carbon (DOC) in the river and the interstitial (int.), total nitrogen (TN) in IS and SS, nitrate (NO₃) in the river and interstitial, C/N atomic ratio, $\delta^{13}C_{org}$, $\delta^{13}C_{tot}$ and $\delta^{15}N$ 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 (%)	0.40.1 (18)	0.5±0.3 (16)	0.5±0.2 (12)
NO_3 river (mg l ⁻¹)	5.2±0.7 (7)	8.9±0.6 (7)	9.0±1.2 (7)
NO_3 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)
$\delta^{13}C_{org}SS(\%)$	-28.0±0.5 (18)	-28.1±0.9 (16)	-27.8±0.9 (15)
$\delta^{13}C_{tot}$ SS (‰)	-20.1±2.8 (18)	-21.8±2.6 (16)	-22.9±3.0 (12)
δ^{15} N SS (‰)	-0.4±0.9 (18)	2.0±1.1 (16)	1.8±0.6 (12)



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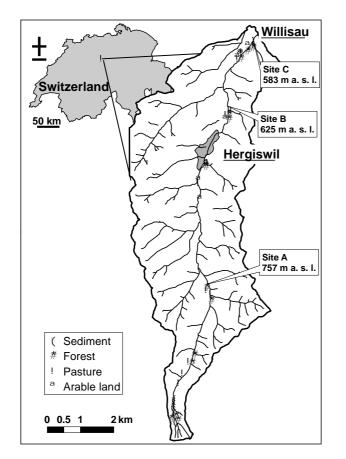
Table 2. Range (mean) of C/N atomic ratio, $\delta^{13}C_{org}$, $\delta^{13}C_{tot}$ and $\delta^{15}N$ of algae, manure and riverbed sediment (riverbed S) as well as of forest, pasture and arable land soils of the watershed.

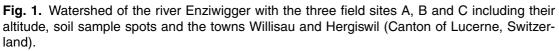
	C/N atomic ratio	$\delta^{13}C_{org}$ (‰)	$\delta^{13} C_{tot}$ (‰)	δ^{15} N (‰)
Algae (<i>n</i> = 4)	7.9–9.6 (9.1)	-41.431.1 (-35.0)	-40.1-16.7 (-24.3)	-0.7-3.5 (2.1)
Manure $(n = 6)$	14.6-34.9 (20.8)	-28.725.3 (-27.9)	-29.025.7 (-27.9)	7.4-11.9 (8.9)
Riverbed S $(n = 5)$	12.1-25. 5 (17.8)	-28.126.7 (-27.3)	-3.20.7 (-1.9)	-5.93.44 (-4.7)
Forest $(n = 14)$	13.4–31.8 (16.8)	-34.826.8 (-28.8)	-28.711.0 (-23.0)	-4.0-4.7 (-0.8)
Pasture ($n = 12$)	11.4-26.1 (13.9)	-29.227.8 (-28.6)	-29.520.7 (-27.6)	-1.0-6.4 (3.0)
Arable land $(n = 8)$	9.5–11.3 (10.7)	-28.527.1 (-27.8)	-28.319.9 (-25.9)	4.3-7.7 (6.0)

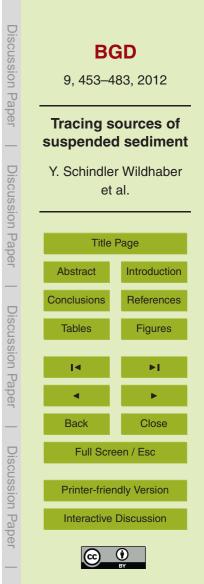
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Table 3. Mean (range \pm sd) source contribution (%) to the suspended sediment at the three sites.

Source	Site A	Site B	Site C
Sediment	$27.0(11.0\pm0.7-52.8\pm0.8)$	18.3 (3.1±2.0-41.2±1.5)	$12.1 (2.0 \pm 1.5 - 48.2 \pm 0.8)$
Forest	$16.0 (0 \pm 1.7 - 45 \pm 2.2)$	16.8 (2.5±1.9–49.1±18.5)	26.4 (3.6±3.1–40.9±12.4)
Pasture	$56.9(28.4 \pm 1.7 - 75 \pm 1.0)$	$25.1(3.8\pm3.4-64.0\pm14.6)$	38.0 (6.7±4.6-56.0±20.7)
Arable land		$39.9(11.7\pm5.1-70.4\pm2.4)$	23.5 (7.2±4.5–45.8±7.7)







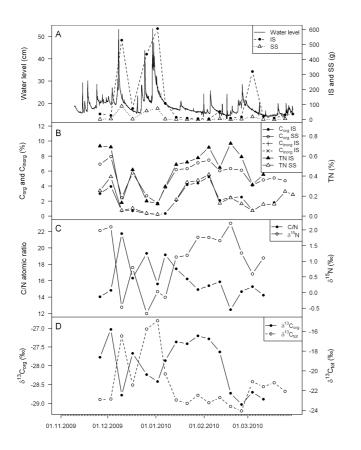
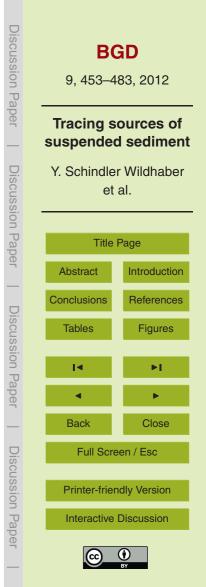
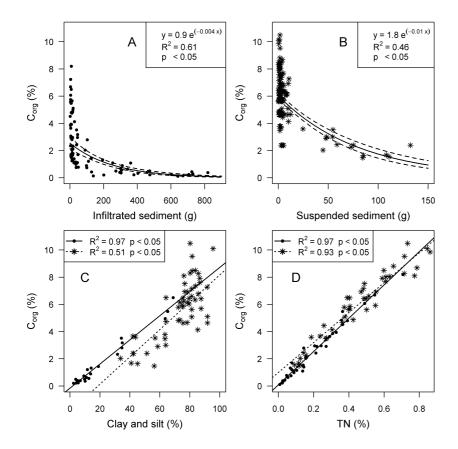


Fig. 2. Sediment, nutrient and isotope dynamics during the field period. Plotted are mean values of all samples. **(A)** Water level at site B, the weekly infiltration of sediment < 2 mm (IS) and the weekly suspended sediment (SS); **(B)** C_{org} and TN of IS and SS; **(C)** C/N atomic ratio and δ^{15}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





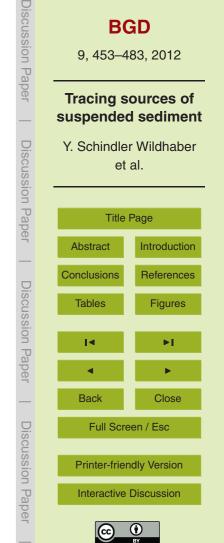
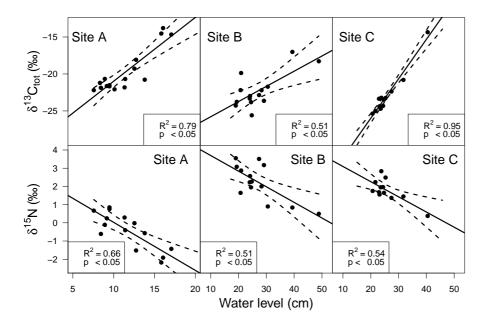
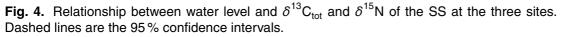
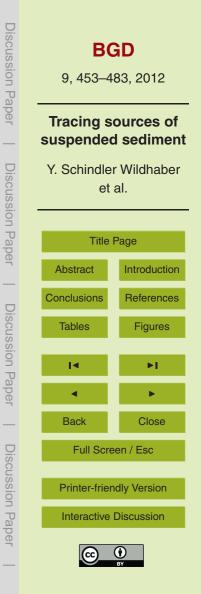


Fig. 3. Relationship between C_{org} and **(A)** total IS < 2 mm; **(B)** total SS; **(C)** clay and silt fraction of sediment < 2 mm and **(D)** TN. Solid circles and solid lines: Infiltrated sediment, stars and dotted lines: SS. Dashed lines in **(A)** and **(B)** are the 95% confidence intervals.







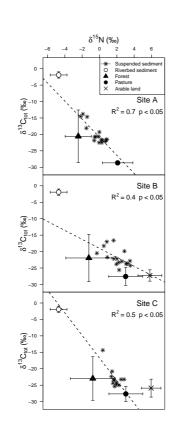
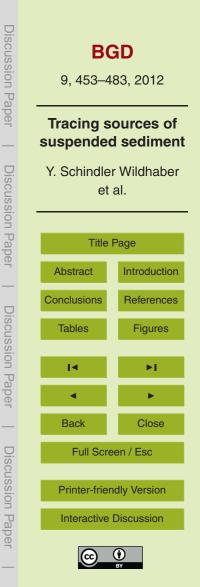


Fig. 5. $\delta^{13}C_{tot}$ and $\delta^{15}N$ of SS and soil samples collected above each sites (average ± sd). Dashed line: SS regression line.



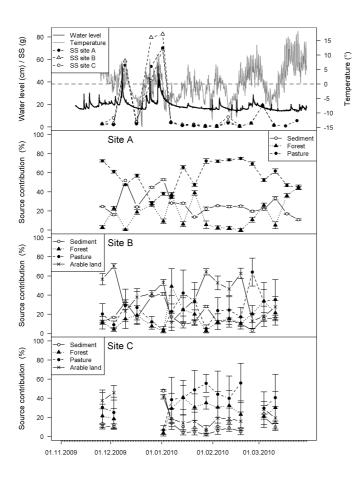


Fig. 6. Total suspended sediment (SS) per week at the three sites and soil source contribution from the three/four possible sources to SS at the three sites, determined with the dual isotope mixing model *IsoSource*. Air temperature was measured close to site B in Hergiswil.

