

# Water Resources Research



# RESEARCH ARTICLE

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#### **Key Points:**

- In over 90% of the cases, gross exchange equals five times the net exchange, which impacts interpretations of nutrient uptake
- Gross exchange and hydrologic turnover show spatiotemporal patterns persisting over discharge at forested, but not at agricultural sites
- Moderate discharge exhibits the highest relative gross exchange

#### **Supporting Information:**

Supporting Information may be found in the online version of this article.

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# Hydrologic Turnover Matters — Gross Gains and Losses of Six First-Order Streams Across Contrasting Landscapes and Flow Regimes

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**Abstract** Gross gains and losses of stream water and the consequent hydrologic turnover may modify the composition of stream water and drive in-stream ecological functioning. We evaluated over 500 breakthrough curves of conservative tracer additions to analyze the channel water balance resulting in gross gains and losses, net exchange, and hydrologic turnover. During the hydrological year 2019, seven tracer experiments had been carried out in six first-order streams along 400 m study reaches. All streams are located in the Holtemme catchment (Central Germany) with three each dominated by forested and agricultural land use. Four of the six streams were characterized by net-losing conditions. The overall median of gross exchange was five times higher than net exchange. On average, subsurface gains replaced 50% of the original stream water over less than one kilometer of stream length. We even observed cases where over 95% of the stream water turned over within 100 m. Gross exchange was relatively higher in forested than in agricultural streams. Patterns of exchange in the forested streams persisted spatially and were temporally independent of streamflow, whereas in the agricultural ones, variable spatial patterns and streamflow dependence occurred. Overall, moderate flow coincided with highest relative gross exchange. Our results support previous findings that in-stream solute concentrations could heavily depend on location and magnitude of gains and losses. Gross exchange embodies a permanent but variable control of downstream solute concentrations interacting with the signal of biogeochemical activity. We highlight the importance to include reach-scale hydrological processes in studies on nutrient spiraling.

Plain Language Summary The vitality of stream ecosystems largely relies on the exchange of water between surface and groundwater. This comprises all gains and losses of stream water from and to the subsurface and is referred to as gross exchange. We investigated gross exchange for six headwater streams in the Holtemme catchment (Central Germany) during the hydrological year 2019. By applying salt tracer experiments we calculated the extent of exchange. Consistently, the investigated stream reaches lost more water than they gained. On average, half of the stream water was replaced by newly added groundwater along less than one km of stream length and, in few cases, almost the entire volume was exchanged within 100 m distance. Streams surrounded by forest exchanged more water than streams in agricultural landscapes. The location and direction of exchange remained similar in the forested streams, but varied temporarily for the agricultural streams. We could show that groundwater represents an important volume of our streams and that the true gross exchange can easily be underestimated if only the sum of gains and losses is measured. Therefore, solute concentrations can be strongly modified by gross exchange, which is important to better understand the transport of solutes in streams.

# 1. Introduction

The exchange of stream water with groundwater, and vice versa, is an important driver of stream ecosystem functioning, such as ecosystem metabolism and solute processing (e.g., Bernhardt et al., 2018; Hall & Tank, 2005; Harvey & Gooseff, 2015; Odum, 1956; Ranalli & Macalady, 2010; Stream Solute Workshop, 1990; Wondzell, 2011). Exchange flows can follow complex pathways generated by a range of in-stream morphological structures from dunes, pool-riffle sequences, in-stream bars to meanders (Kasahara & Wondzell, 2003; Trauth et al., 2014, 2015). In addition, heterogeneous permeability structures control the magnitude and patterns of exchange flow (Bergstrom et al., 2016; Covino & McGlynn, 2007; Fleckenstein et al., 2006; Irvine et al., 2012;



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Kalbus et al., 2009). Similarly, water exchange is influenced by spatially smaller and temporally more variable biological components such as woody debris and leaf litter, macrophyte or biofilm growth, and bioturbation (Battin et al., 2008, 2016; Hart et al., 1999; Mermillod-Blondin, 2011; Mutz et al., 2007; Nogaro et al., 2006). Sediment permeability can be further modified by physical or biological clogging through deposition of fine sediment or organic matter (Beschta & Jackson, 1979; Hartwig & Borchardt, 2015; Nogaro et al., 2010; Schälchli, 1992). At larger scales, geomorphological relationships suggest that streamflow increases downstream with increasing contributing area (Rodríguez-Iturbe & Rinaldo, 1997), yet many streams and rivers lose water locally (Jasechko et al., 2021).

Exchange flows, regardless of their scale, for example, for entire stream networks or for stream reaches, cannot be observed directly. Instead, solute tracer tests are widely applied to quantify the integrated effects. Exchange flows that occur at smaller spatial and shorter temporal scales (also conceptualized as transient storage) are encoded in the tailing of a tracer breakthrough curve (BTC). Losing fluxes and storage on time scales longer than the tracer experiment can be detected as tracer mass that is not recovered within the BTC and is "lost" into the subsurface. By incorporating this information into the channel water balance, both net and gross changes in discharge can be estimated. Previous studies have already shown that surface-subsurface exchange is a bidirectional process, with gross losses and gains co-occurring over short distances at reach scale (e.g., Payn et al., 2009; Ruehl et al., 2006; Valett et al., 1996). Gross gains and losses modify the stream water composition even if net changes in streamflow are small. The fraction of water, which has entered the channel at a certain location, will decay during the travel downstream. This evolution of stream water composition has been termed fractional hydrologic turnover (Covino & McGlynn, 2007). Covino et al. (2011) define hydrologic turnover and the associated changes in stream water composition "as the process of streams losing a fraction of water to groundwater while sequentially or simultaneously receiving different water from groundwater." Therefore, understanding hydrologic turnover has profound implications for the transport of solutes in streams and their fate in the channel network (Mallard et al., 2014; Payn et al., 2009). Furthermore, Ward et al. (2013) note the importance of combining transient storage with a mass balance approach to better understand solute transport and processing. Water gained after short storage will most likely have a similar chemical composition, while gains of older groundwater may be chemically distinct from stream water (Batlle-Aguilar et al., 2014; Lee et al., 2020). Thus, mixing of water with different concentrations of reactive solutes (e.g., nitrate) and hydrologic turnover interfere with patterns of solute reactions such as denitrification or nutrient uptake.

To date, most studies of nutrient processing have conceptualized stream-subsurface interaction from a stream perspective where stream water, which has entered the subsurface, re-emerges into the stream after some travel time and flow through storage zones (Bencala et al., 1983; Bencala & Walters, 1983; Newbold et al., 1981; Runkel & Bencala, 1995). Such short-term, transient storage, facilitated by hyporheic exchange, does not modify the mass balance of the stream. Changes in solute concentrations are predominantly attributed to biogeochemical processes and additionally approximated by changes in the apparent lateral in- and outflow, which are net changes in discharge between observation sites, known groundwater sources, drainages, and tributaries (Roberts & Mulholland, 2007; Runkel, 1998; von Schiller et al., 2011). Gross gains and losses and the resulting hydrologic turnover may very likely alter in-stream nutrient concentrations beyond apparent lateral flow paths (Harvey et al., 2019). These mechanisms, however, are currently not well represented in conceptualizations of nutrient uptake and nutrient spiraling.

A variety of factors influence the magnitude and spatial patterns of hydrologic turnover. Several studies have examined the impacts of hydrologic turnover driven by network geometry, stream order, topography, and geology on stream channel water balance (e.g., Bergstrom, Jencso, et al., 2016; Covino et al., 2011; Haggerty et al., 2002; Valett et al., 1996; Ward et al., 2019; Wondzell, 2011). The role of temporal changes accounting for variability with flow regime has also been investigated, but these studies tended to focus on shorter time scales or only on transient storage (Payn et al., 2012; Ward et al., 2018a; Ward, Gooseff, et al., 2013; Zarnetske et al., 2007). The interplay of different spatial and hydrologic drivers has been studied (Schmadel et al., 2017; Ward et al., 2018b). Moreover, existing research suggests that gross gains are associated with higher levels of vegetation, particularly during base flow (Bergstrom et al., 2016). Nevertheless, a systematic understanding of factors that influence gross gains and losses in time and space is currently missing.

In this study, we quantified gross gains and losses as well as hydrologic turnover based on evaluations of conservative tracer BTCs for six streams in central Germany. Three streams were less anthropogenically impacted forest

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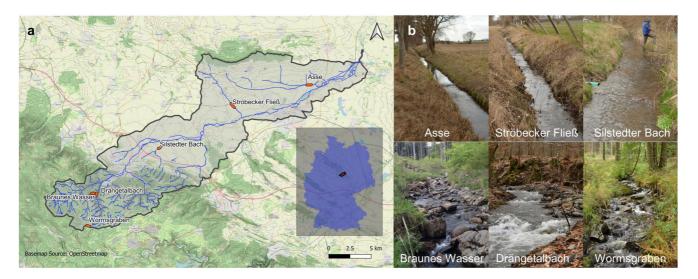


Figure 1. (a) The Holtemme Catchment is located in Central Germany. Red dots represent sites at the six streams of this study. The topographic base map roughly distinguishes forested areas (greenish) and agricultural land (yellowish). (b) Representative reaches of the three agricultural (upper panels) and three forested streams (lower panels): Asse, Ströbecker Fließ, Silstedter Bach, Braunes Wasser, Drängetalbach, Wormsgraben.

streams, and the other three were heavily impacted agricultural streams. Seven tracer experiments were conducted in each stream at several times of the year and at different discharge conditions. We performed a detailed analysis of the extensive data set, which includes over 500 BTCs, addressing the following questions:

- 1. Which is the magnitude of gross gains and losses, and are there differences among land use types?
- 2. Can we identify spatial patterns of gross exchange as well as hydrologic turnover and their driving forces? Which is the larger control: geomorphology or streamflow (Schmadel et al., 2017; Ward et al., 2018b)?

Our key motivation was to corroborate the notion that gross gains and losses and thus, hydrologic turnover may substantially alter stream water composition and need to be considered when assessing in-stream nutrient processing.

#### 2. Methods

We collected data slightly beyond the hydrological year of 2019 (from September 2018 to October 2019) in six first-order streams located within the Holtemme catchment in Central Germany (Figure 1a). Monitoring included seven campaigns at each stream with conservative (NaCl<sup>-</sup>) tracer experiments. These campaigns covered different flow conditions and seasons. From the experiments, we calculated a stream channel water balance per stream and campaign. We used those results to obtain a measure of hydrologic turnover and analyze spatiotemporal patterns within and amongst the streams.

# 2.1. Study Area

The Holtemme catchment (282 km², Mueller et al., 2016) covers different topographic and geologic features. The Northern Harz margin fault roughly divides the catchment at about the city of Wernigerode (Figure 1a) into the mountainous Harz region (1141 m a.s.l.), which is dominated by the Palaeozoic granite intrusion (283 Ma) surrounded by Devonian schist and greywacke as bedrocks (Baumann et al., 1991; Zech et al., 2010), and the lowlands of Magdeburger Börde with their carbonic sediments and fertile black soils (Schubert, 2008). The six streams of our study (Figure 1a) align along the topographic gradient, which is reflected in stream characteristics and the gradient of land use alike (Weitere et al., 2021): starting in the mainly coniferous forest Harz regions in the West (Wormsgraben/WoG, Braunes Wasser/BrW, Drängetalbach/DrB—hereafter forested streams), continuing to the lowland, agricultural plains in the East with some urban areas (Silstedter Bach/SiB, Ströbecker Fließ/StF, Asse—hereafter agricultural streams). The catchment size of the forested streams is generally one order of magnitude lower than that of the agricultural streams (average 4.4 vs. 25.7 km², Table S1 in Supporting

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Information S1). Hydromorphologically, the forested streams have channel morphologic characteristics with steeper slopes, pool-riffle sequences, in-stream gravel bars, meandering glides or small falls, and accommodate woody debris (Figure 1b). Though, one of the forested streams, WoG, was constructed as a ditch for timber floating. Therefore, the WoG stream bed mainly consists of bedrock and small drainages, which come from meadows used for pasture farming during parts of the year, flow into the downstream part of the reach. The agricultural streams of StF and Asse are highly impacted lowland streams: straightened and incised, having low slopes, some widening and narrowing of the stream bed with minor riffles, and some pool-like structures (Figure 1b). The stream of SiB drains the transitional zone close to the fault and is characterized by agriculturally dominated land use but shows more hydromorphological variability than the latter two. In conclusion, the different stream types, nutrient levels (Pottgiesser, 2018), and grain size distributions (Table S2 in Supporting Information S1, pers. comm. J. Pasqualini) of the six streams support the idea of a gradient and thus, a grouping into forested and agricultural land use (Table S1 in Supporting Information S1).

# 2.2. Monitoring Setup

During each sampling campaign (Table S3 in Supporting Information S1), a reach of 400 m per stream was sampled at five sites, that is, every 100 m, identical throughout the monitoring. Sampling covered the following field parameters: wetted width (ww in cm), specific electrical conductivity (EC in  $\mu$ S cm<sup>-1</sup>), temperature (T in °C), and discharge (Q in L s<sup>-1</sup>) at the most downstream site (400 m).

Wetted width was measured three times at each site in a range of roughly one meter up/downstream of the site. Accuracy here accounts for  $\pm 1$  cm. Discharge was measured with an MF Pro Otis device (OTT HydroMet GmbH, accuracy  $\pm 2\%$  of reading). EC and T were logged at all five sites of the reach during a tracer experiment with a resolution of 5 s intervals using either CTD-Diver (Eijkelkamp Soil & Water, accuracy: Conductivity  $\pm 1\%$  of reading, pressure  $\pm 0.5$  cm  $H_2O$ , Temperature  $\pm 0.1$ °C) or LTC Levellogger Edge (Solinst Canada Ltd, accuracy: Conductivity  $\pm 2\%$  of reading, pressure  $\pm 0.05$  cm  $H_2O$ , Temperature  $\pm 0.05$ °C). For all sensors, individual EC—Cl<sup>-</sup> calibration curves were obtained prior to the first set of tracer experiments.

BTCs represent the integrated effects of solute transport and storage processes at the reach scale, which are downstream advection, longitudinal dispersion, and transient storage (Schmadel et al., 2016). It is inherent to tracer experiments that storage longer than the duration of the experiment cannot be measured directly; in other words, experiments longer than the tailing of BTCs where the tracer is still detectable. The logging period during our experiments was sufficiently long to capture the full BTC from first arrival to the tailing. We varied logging according to the reach length (single reach or combined reaches) between 4 and 48 hr. To calculate the channel water balance in our study, we focus on the analysis of tracer recovery and the tracer mass that was not recovered during the logging period.

# 2.3. Stream Channel Water Balance

Based on the principles of dilution gauging (Day, 1976) and mass recovery (Harvey et al., 1996; Triska et al., 1989), we performed an instantaneous injection of dissolved NaCl<sup>-</sup> tracer every 100 m along the study reaches. Starting at the most downstream site to avoid overlap of the injections, we obtained a maximum of 15 BTCs per stream and campaign. The amount of added salt (25-2500 g) was estimated by simulating BTCs with an analytical solution of the advection-dispersion equation and optimize the added tracer mass for peak concentrations of EC to be 1.5 times higher than the background at the most downstream site after the last injection 400 m upstream (ideally even less, i.e.,  $100 \text{ mg L}^{-1}$  in agricultural and  $20 \text{ mg L}^{-1}$  Cl in forested streams). Mixing length, the distance between site of tracer injection and start of the stream reach, was estimated each time visually. We followed the rule to capture at least three successive morphological changes within this distance, for example, three sequential riffles or widening/narrowing of the stream (Payn et al., 2009), with a minimum of 10 m length (median  $13 \pm 3$  m in this study).

The first measured BTC at each site was used to estimate discharge, assuming that over the mixing length no tracer mass would be lost under constant discharge conditions (Kilpatrick & Cobb, 1985):

$$Q_i = \frac{M_{init}}{\int_0^t C_i(t)dt} \tag{1}$$

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where  $Q_i$  is discharge,  $M_{init}$  the initial tracer mass injected, and  $C_i(t)$  the integrated tracer  $Cl^-$  concentration below the BTC. We then used this discharge to calculate the net change in discharge along the stream subreaches:

$$\Delta Q = Q_i - Q_{i-1} \tag{2}$$

To obtain hydrologic gross gains and losses (as in Covino et al., 2011; Payn et al., 2009), we calculated lost tracer mass  $(M_{loss})$  as the difference between recovered mass  $(M_{rec})$  downstream and initially injected mass upstream  $(M_{init})$ :

$$M_{Loss} = M_{rec} - M_{init} = Q_i \int_0^t C_{(i-1)D}(t)dt - M_{init}$$
 (3)

with  $Q_i$  discharge downstream and  $C_{(i-1)D}(t)$  the upstream tracer concentration logged downstream.  $M_{loss}$  becomes negative as soon as a fraction of injected mass is lost over the reach. The corresponding gross loss  $(Q_{loss})$  was calculated as:

$$Q_{loss} = \frac{M_{loss}}{\int_0^t C_{(i-1)D}(t)dt}$$
(4)

 $M_{loss}$  is dependent on the in-stream tracer concentration, as pointed out by Payn et al. (2009), and  $Q_{loss}$  will be potentially impacted by gaining water (tracer dilution/enrichment along the reach). Using the calculation for  $Q_{loss}$  as in Equation 4, we have assumed a maximum dilution before loss (gain before loss) for our results of the channel water balance.

The sum of gross gain ( $Q_{gain}$ ) and  $Q_{loss}$  would be equivalent to the net change in Q ( $\Delta Q$ ), accordingly:

$$\Delta Q = Q_{gain} + Q_{loss} \tag{5}$$

where  $Q_{gain}$  is positive and  $Q_{loss}$  is negative.

In contrast to net change, which will be large if loss and gain are very distinct and small if they are of similar magnitude, gross exchange  $Q_{gross}$  is represented by the sum of absolute values of  $Q_{loss}$  and  $Q_{gain}$ .

$$Q_{gross} = |Q_{loss}| + Q_{gain} \tag{6}$$

Consequently, gross exchange will always exceed net exchange, except in cases where either gross gain or loss is zero. This would result in parity of net and gross exchange. The difference between  $Q_{gross}$  and  $\Delta Q$  is maximal when  $Q_{gain}$  equals  $Q_{loss}$ , which results in  $\Delta Q$  of zero. For better comparison of  $Q_{gross}$  and net exchange, we further define net exchange  $Q_{net}$  as the absolute values of  $\Delta Q$ .

Prior to all calculations, we corrected EC data for background conductivity, applying an asymmetric least-square fit to subtract background levels (R package *baseline*, Liland et al., 2010). All BTCs were separated, whereby overlapping ones would be truncated. Furthermore, BTCs were classified into three categories: (a) *low quality* regarding signal-to-noise levels, incompleteness, measurement failure or overlap, (b) *intermediate to good quality* mainly owing to a slight overlap of consecutive additions at the end of BTC tail, and (c) *very good quality* without concerning issues.

For the integration of the BTCs, we applied trapezoidal numerical approximation of the EC below the BTC. The start of the integration window was chosen visually, whereas the end was reached as soon as the value of EC again stabilized around the starting value  $\pm 0.5~\mu S$  cm<sup>-1</sup>. Many studies before have limited their window of detection and truncated the BTC to  $t_{99}$  (99% of BTC passed) to reduce error regarding the signal-to-noise interferences in the long tailing (Schmadel et al., 2016; Ward, Gooseff, et al., 2013; Ward, Payn, et al., 2013). In our study, we compared outcomes for a window of detection between  $t_{85}$  and  $t_{100}$  (Table S4 in Supporting Information S1) but used the entire integration time  $t_{100}$  for further analysis.

We validated the mass balance approach by checking the data quality in two ways: (a) we excluded mass recoveries of the injected tracer mass greater than 100% from the calculation of the stream channel water balances, and (b) adjusted cases in which we found physically impossible outcomes of positive losses or negative gains. For the latter, we zeroed the incorrect losses/gains and set the associated gains/losses to the value of net exchange

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between the sites (as suggested by Payn et al., 2009). Furthermore, we normalized all gross losses and gains by stream discharge to make outcomes comparable between the individual streams, and the term relative exchange refers to such normalized values.

#### 2.4. Hydrologic Turnover and Spatiotemporal Patterns

Concurrent gross gains and losses result in an evolution of the source composition of stream water, which has been termed fractional hydrologic turnover by Covino et al. (2011). Here, we apply the concept of hydrologic turnover to understand the spatial development of gross gains and losses of water from the stream to the groundwater and vice versa along the stream reach (Covino et al., 2011; Payn et al., 2009).

As gains and losses at a stream reach occur simultaneously and cannot be further spatially separated, the quantification of hydrologic turnover can be based on the following two assumptions: (a)  $Q_{gain}$  enters the stream reach and then, a fraction of the stream water residing in the stream  $(Q_{i-1})$  as well as the water gain are lost (gain before loss) or (b) a fraction of  $Q_{i-1}$  that enters the actual reach is lost and then  $Q_{gain}$  is added (loss before gain). Mallard et al. (2014) quantified hydrologic turnover based on the gain before loss assumption, while we applied the loss before gain case. We estimated a fraction remaining  $(F_{rem})$  of upstream water entering the next 100 m subreach along the 400 m study reach by:

$$F_{rem} = 1 - \frac{Q_{loss,i}}{Q_{i-1}} \tag{7}$$

Correspondingly, 1 L lost over 100 m with an initial upstream discharge of 10 L would than result in 9 L remaining for the next 100 m reach, yielding a  $F_{rem}$  of 0.9. The results for  $F_{rem}$  indicate the magnitude of hydrologic turnover in the subreach, in other words, the smaller the fraction the more water has been replaced within a 100 m distance. Accordingly, the  $F_{rem}$  in the current reach i from the jth upstream reach is the product of all  $F_{rem}$  from reach j to reach i:

$$F_{rem_{i,j}} = \prod_{i=1}^{j} F_{rem_i} \tag{8}$$

If there was a lack of data for one subreach, we assumed a  $F_{rem}$  of 1, so no loss occurring within those 100 m. In this way, we could calculate the remaining volume of upstream water in the downstream reaches by multiplying  $F_{rem}$  with the upstream discharge. The approach of loss before gain enabled us to estimate hydrologic turnover in a more conservative way, that is, lower turnover with higher values of  $F_{rem}$  compared to the assumption of gain before loss

Additionally, to better demonstrate the effect of hydrologic turnover in the individual streams, we calculated turnover lengths based on medians of  $F_{rem}$  per stream. We considered turnover lengths for the distances after which 50% or 10% of the upstream water had been lost to the subsurface and replaced by new water. For providing a robust comparison of the spatiotemporal patterns of gross exchange and hydrologic turnover, we applied a rank-based assessment in addition to the data at the metric scale. We chose to rank the magnitude of exchange (per subreach) and search for the frequency of the highest exchange/turnover among the campaigns. Besides  $F_{rem}$ , we applied this to all parts of the stream channel water balance: net and gross exchange, gross loss, and gain. Beforehand, values were normalized by the corresponding discharge to obtain a comparable range for ranking the data.

#### 3. Results

# 3.1. Stream Channel Water Balance

The seven campaigns for each of the six streams consisted of five consecutive NaCl<sup>-</sup> tracer additions every 100 m along the study reach resulting in 548 breakthrough curves (BTCs) (see Table S3 in Supporting Information S1 for missing BTCs due to temporally limited access at Braunes Wasser: 1/7 and flow intermittence at Ströbecker Fließ: 3/7). We categorized BTCs into groups of (a) *low quality* (30 BTCs, 5.5%), (b) *intermediate to good quality* (229 BTCs, 41,8%), and (c) *very good quality* (289 BTCs, 52,7%) based on the criteria listed in the Methods section and could use 513 BTCs for integration (35 BTCs removed out of categories 1 and 2). Afterward, we excluded mass recovery rates larger than 100% from further processing (61 BTCs, 11%). In

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**Table 1** Summary of Mass Balance Results per Individual Stream Over All Reaches and Campaigns Presented for the Parameters: Discharge Q, Mass Recovery  $M_{rec}$ , Gross Loss  $Q_{loss}$ , Gross Gain  $Q_{gain}$ , Net Exchange ( $\Delta Q$  See Equation 2), and Gross Exchange ( $Q_{gross}$ , See Equation 6)

Parameter	Stream	Mean	Median	Min	Max
Q [L s <sup>-1</sup> ]	Asse	23.5	21.8	4.3	59.1
	StF	10.0	8.2	0.8	24.4
	SiB	19.1	17.8	10.9	40.8
	BrW	32.8	18.2	4.9	110.9
	DrB	42.1	20.4	4.7	196.3
	WoG	14.9	11.0	2.7	41.7
$ m M_{rec}~\%$	Asse	84.6	88.2	55.8	99.5
	StF	82.0	83.5	50.5	99.9
	SiB	84.3	89.2	50.2	99.8
	BrW	73.1	78.1	32.7	99.1
	DrB	73.6	78.9	25.9	99.1
	WoG	89.8	92.9	45.0	100.0
$Q_{loss}[L\;s^{-1}]$	Asse	-4.9	-3.3	-21.0	-0.2
	StF	-1.9	-1.1	-5.4	0.0
	SiB	-3.5	-1.6	-22.5	0.0
	BrW	10.4	-4.2	-65.9	0.0
	DrB	-8.5	-5.3	-45.9	-0.3
	WoG	-1.6	-1.4	-11.2	0.0
$Q_{gain}[Ls^{-1}]$	Asse	3.5	1.8	0.0	18.4
	StF	1.3	0.2	0.0	7.7
	SiB	2.8	2.0	0.0	12.0
	BrW	5.7	3.2	0.0	39.0
	DrB	9.7	6.6	0.0	45.7
	WoG	1.6	0.9	0.0	8.1
$\Delta Q [L s^{-1}]$	Asse	-1.4	-0.8	-21.0	15.0
	StF	-0.6	-0.1	-5.3	5.6
	SiB	-0.7	-0.2	-22.5	4.1
	BrW	-4.4	-0.2	-49.8	21.8
	DrB	1.2	1.0	-27.4	23.1
	WoG	0.0	0.1	-11.2	5.6
$Q_{gross}[L\;s^{-1}]$	Asse	8.4	4.3	0.6	29.0
	StF	3.1	2.1	0.0	11.1
	SiB	6.2	4.0	0.9	28.1
	BrW	16.1	7.5	1.4	88.4
	DrB	18.2	11.6	0.8	68.3
	WoG	3.2	1.9	0.1	11.2

Note. Agricultural: Asse, StF, SiB; Forested: BrW, DrB, WoG.

general, BTCs with such unrealistic mass recoveries comprised additions in the agricultural streams with high and noisy background EC levels or overall conditions of higher discharge regime. Finally, we calculated channel water mass balances from 452 BTCs. In 39 cases (8.6%), we set gross exchange equal to net exchange for physically not meaningful results of  $Q_{loss} > 0$  and  $Q_{gain} < 0$ . Data for this study, including channel water balance and results of hydrologic turnover, are available in a repository (Jähkel, 2021).

Across all streams, discharge covered three orders of magnitude (median 16,5 L s<sup>-1</sup>, Table 1) ranging from 0,8 L s<sup>-1</sup> during base flow at StF to 196 L s<sup>-1</sup> during a winter campaign at DrB. We found the highest absolute mean values for Q<sub>loss</sub> and Q<sub>gain</sub> in two forested streams—BrW and DrB (means Table 1, relative medians Figures 2a and 2b). The maximum gross exchange of 88.4 L s<sup>-1</sup> was observed at BrW, corresponding to more than two-thirds of the discharge at the downstream end of the 100 m reach. Such cases of high exchange, where gross exchange exceeded 50% of the downstream discharge, accounted for 15% of the data and occurred in all streams but most frequently in DrB and BrW. In addition, gross exchange was always substantially greater than net exchange: (a) for the entire data set (overall means: net  $-1 L s^{-1} vs$ . gross 10.6 L s<sup>-1</sup>, Table 1) and (b) for individual streams (medians of relative gross exchange 4-38 times larger than those of relative net exchange, Figure 2c vs. Figure 2d). The exceptions noted above included about 9% of the data where net exchange equaled gross exchange, because of otherwise unrealistic results. In summary, four of the six streams were on average net-losing, and the overall lost tracer mass was 19% (Table 1).

Median relative gross exchange was nearly twice as high in forested streams BrW and DrB than in agricultural streams (46 and 49,3% of Q vs. 23%,5%–30% of Q, Figure 2d). Results for WoG were exceptionally low (median 17,8%). Additionally, we observed a tendency for higher variability of net exchange in agricultural than in forested streams (Figure 2c). However, net exchange was low for all streams compared to gross exchange, and again, WoG had the lowest values — confined to zero. Despite their overall lower medians, agricultural streams also exhibited variability in gross losses and gains (Figures 2a and 2b).

#### 3.2. Hydrologic Turnover

Hydrologic turnover was calculated based on  $F_{\rm rem}$  — the fraction remaining of upstream water entering the next subreach — and its cumulative products for the various subreaches along the 400 m study reach. Hydrologic turnover increases with decreasing  $F_{\rm rem}$ . The overall average of  $F_{\rm rem}$  was 0.79, equivalent to 21% of water exchanged along the study reaches (Figure 3a). The highest median values of  $F_{\rm rem}$  occurred at SiB and WoG (0.85 and 0.88), whereas the lowest values were observed at DrB and BrW (0.63 and 0.75, Figure 3a). Thus, with the exception of WoG, a lower  $F_{\rm rem}$  and thus, a higher hydrologic turnover was observed for the forested streams than for the agricultural streams. These results of exchange are supported by the short turnover lengths of 600 and 700 m (DrB, BrW) compared to longer 1100–1500 m distances (SiB, WoG) after which 50% of the water would have been replaced (Figure 3b). Notably, the lowest values of  $F_{\rm rem}$  reflected almost a 100%

exchange along 100 m were observed at DrB ( $F_{rem}$  0.02) during summer and autumn campaigns. About one percent of our data showed such a high exchange (>95%) over a distance of 100 m.

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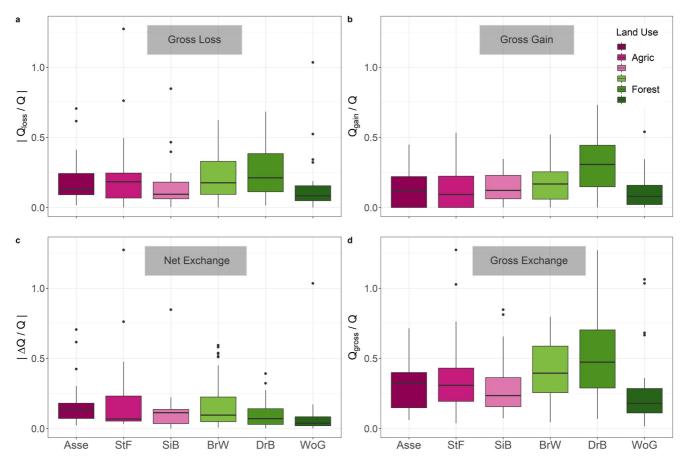


Figure 2. Results for main components of channel water balance per individual stream, normalized by local discharge: Relative (a) Gross Loss —  $Q_{loss}$  (absolute values), (b) Gross Gain —  $Q_{gain}$ , (c) Net exchange  $\Delta Q$  — net change between up- and downstream reaches (absolute values), and relative (d) Gross Exchange  $Q_{gross}$  — the sum of losses (absolute values) and gains.

In summary, we consistently observed high turnover and, consequently, short turnover lengths (almost half the distance, Figure 3b), in two of the forested streams, in particular, while hydrologic turnover was lower in the other streams.

#### 3.3. Spatiotemporal Patterns of Gross Exchange and Hydrologic Turnover

In general, the streams exhibited individual patterns of hydrologic turnover along the reaches over time (Figure 4). In DrB and BrW, hydrologic turnover patterns remained similar throughout the year, that is, losing/gaining reaches were losing/gaining at all times but differed in magnitude (overall negative slopes). For the other streams, gross losses and gains varied spatially and temporally, yielding distinct patterns of hydrologic turnover (variable slopes). Because the magnitudes of  $Q_{loss}$  and  $Q_{gain}$  were highly variable spatially and temporally, we present the detailed comparison of spatiotemporal patterns by ranking the variables, which provides a more robust picture.

The ranking of  $Q_{loss}$ ,  $Q_{gain}$ , net exchange ( $Q_{net}$ ), and gross exchange ( $Q_{gross}$ ), as well as  $F_{rem}$ , revealed which reaches were predominantly gaining, losing, or both, that is, showed a characteristic exchange behavior throughout the entire year (Figure 5). Overall, forested streams (lower panels) tended to have more distinct reach characteristics, whereas the ranking of reaches in agricultural streams (upper panels) was more variable over time. Reaches with persistently high ranks were observed in the following streams: BrW subreach 300–400 m (the highest rank for all variables in all campaigns, except  $Q_{gain}$ ), DrB subreach 200–300 m (highest for  $F_{rem}$  86% and  $Q_{loss}/Q_{gain}$  57% of all campaigns), and WoG subreach 100–200 m (highest for all variables but  $Q_{gain}$ ). We found almost no peaks for the agricultural streams but a homogeneous ranking of the parameters in the subreaches. One exception was the most downstream subreach 300–400 m at Asse for  $Q_{loss}$  (highest rank in 71% of all campaigns). In essence, this

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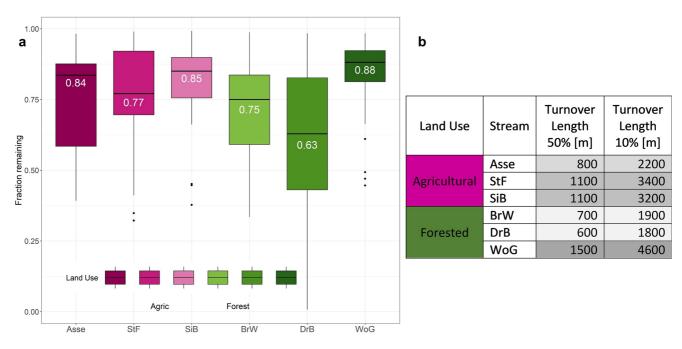


Figure 3. (a) Fractions remaining ( $F_{rem}$ ) for each stream (per reach and products of the subreaches) grouped by land use. Median values are given in the box plot. (b) Turnover lengths for the different streams in meters. Calculations were obtained for 50% and 10% of the remaining upstream water, respectively.

implies that (high) exchange occurred at different sites at different times of the year, as was also indicated by the variable patterns in Figure 4 (upper panels).

In summary, the patterns in the forested streams were persistent for active reaches at different discharge conditions for almost all parameters. In contrast, for agricultural streams, positions of the most active reaches varied over time, and only one or two parameters ranked first in the same subreach.

### 3.4. Temporal Variability of Gross Exchange With Streamflow

In addition to the spatial patterns, a relationship between gross gains and losses and discharge was found (Figure 6). The scatterplots in Figures 6a and 6b show relative  $Q_{loss}$  and  $Q_{gain}$  compared to discharge, colored by the corresponding quartiles of discharge. High relative  $Q_{loss}$  and  $Q_{gain}$  were associated with moderate discharges. High discharge was characterized by low relative values of  $Q_{loss}$  and  $Q_{gain}$ . When aggregated into discharge percentiles and independent of land use, the relative maximum exchange (loss or gain) occurred within the 50th and 75th percentile (Figures 6c and 6d).

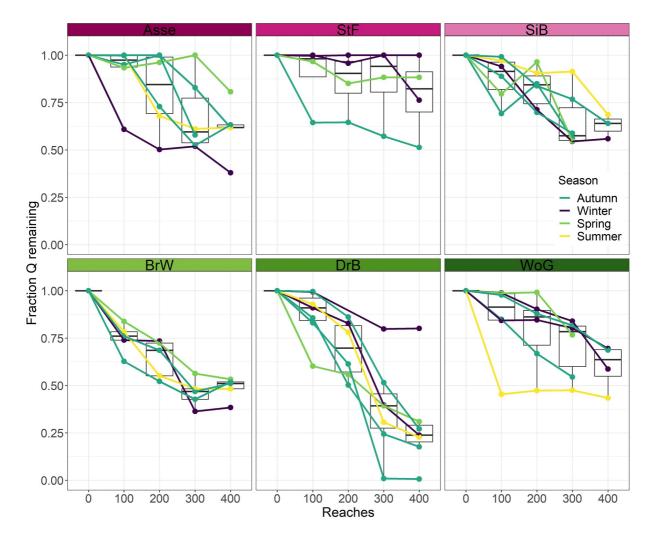
Consequently, moderate discharge appeared to be most important for relative exchange in this study. In particular, the forested streams of DrB and BrW (shape coded in Figures 6a and 6b) were most active for moderate summer and autumn discharges. They were largely inactive during the highest and lowest flows, respectively. In contrast, some agricultural streams reacted on high flow and showed the highest exchange magnitudes at these times, for example, at SiB during spring season (Figure 6a).

# 4. Discussion

# 4.1. The Importance of Gross Exchange and Hydrologic Turnover at the Reach Scale

Our results support the bidirectional nature of gross exchange—losses and gains coincide over short distances as observed in Covino & McGlynn, 2007, Payn et al., 2009 or Zimmer & McGlynn, 2017. Similarly, our results highlight the relevance of gross exchange in terms of exchange flow relative to total stream discharge as well as to net exchange. In 15% of the tracer tests, the magnitude of estimated gross exchange (in absolute values) accounted for more than half of the discharge volume downstream of the reach. On average, gross exchange accounted for five times (median values) the net exchange in our study. We found equality of net and gross exchange in only

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**Figure 4.** Fraction remaining of upstream discharge (site 0 m) at downstream sites along the 400 m reaches for all streams and seasons each (agricultural — upper panels, forested — lower panels). The slope of the curve depicts the magnitude of turnover along the reach, that is, it is high with a negative slope and is low to stagnant when the slope is positive or zero. The box plots depict the variability across all campaigns for each subreach along the study reach.

9% of our data. The high values of hydrologic turnover also indicate the importance of measured gross exchange. Here, we could identify 12%–37% of the water being exchanged along the study reach for most cases (medians of fraction remaining  $F_{rem}$  in Figure 3a), albeit maxima of almost 100% exchange over 100 m were recorded (DrB, Figure 3a).

In general, turnover lengths ranged between 1.8–4.6 km until only 10% of the original upstream water volume remained (Figure 3b). Our estimated turnover lengths agree well with previous studies. In these, nearly all of the upstream volume had been exchanged after one (Covino & McGlynn, 2007) or five (Covino et al., 2011) kilometers. Our results emphasize that hydrologic turnover substantially impacts the source composition of stream water over relatively short distances. Whenever the water gained from the subsurface has a different chemical signature than the stream water, such as in nutrient composition, hydrologic turnover can alter in-stream concentrations that might otherwise be attributed to biogeochemical reactions.

# 4.2. Gross Exchange and Hydrologic Turnover Across Land Use Types and Their Spatiotemporal Patterns

Alongside the general importance of gross exchange at the reach scale, we investigated differences among land use types. We found a more prominent and faster water exchange in the forested compared to the agricultural streams (Figures 2, 3, and 4). However, there was substantial variability between the individual streams and land

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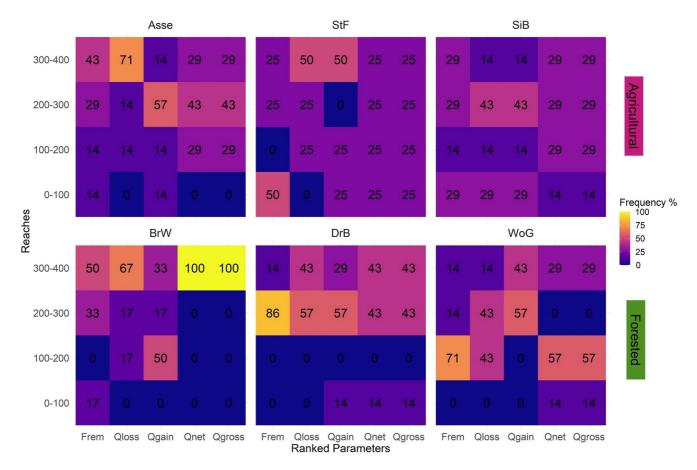


Figure 5. Ranking of the mass balance components for each stream and reach over time: fraction remaining  $F_{rem}$ , gross losses  $Q_{loss}$  and gains  $Q_{gain}$ , net exchange  $Q_{net}$ , and gross exchange  $Q_{gross}$ . The relative frequency of the highest values for each parameter is shown per stream and subreach in percent of the total amount of campaigns, that is, 100% indicates a reach that had the highest value over the entire study period; for  $F_{rem}$  the frequency of the lowest values was used for ranking as this represents the highest turnover.

use types. The forested and the agricultural streams were characterized by different spatiotemporal exchange patterns. The forested, less impacted streams had similar, persistent patterns for all campaigns (consistent slopes in Figure 4) and across all reaches. For example, reaches with high  $Q_{gain}$  tended to have the highest gains (ranking in Figure 5). Exchange flows in the forested streams with their coarse streambed and complex morphological structures seem to be dominated by larger-scale morphological features and subsurface structures that remain independent of flow over extended periods (e.g., Schmadel et al., 2017). There is increasing evidence that general patterns of losing or gaining stream reaches tend to be spatially persistent (Frye, 2013; Ward et al., 2016; Zhang et al., 2021). In contrast, the lowland agricultural streams were characterized by temporarily variable exchange patterns (Figure 5). The higher spatial variability of water exchange is potentially related to seasonal changes in streambed hydraulic conductivity driven by fine sediment inputs through erosion from arable land (Matthaei et al., 2010; Nowinski et al., 2011; Rehg et al., 2005; Schälchli, 1992) and by the growth of benthic biomass or macrophytes (Nogaro et al., 2010; Preiner et al., 2020).

Overall, we argue that the hydrologic patterns found for agricultural and more heavily impacted streams are more dependent on changes in runoff due to their geomorphologic boundary conditions and additional anthropogenic imprinting compared to less impacted forested streams. For example, a constant hydrologic turnover associated with a larger scale morphological feature such as a pool-riffle structure may be overridden by temporary drainage inflow. Therefore, we suppose that geomorphological structure alone cannot entirely explain the differences in gross gains and losses and hydrologic turnover between land use types. We argue that an increasing level of anthropogenic impact goes along with more variable interactions in terms of water exchange. Agricultural streams might become more sensitive to sustained anthropogenic impacts and at the same time, initially dominating geomorphological characteristics might lose their importance. However, where the geomorphological setting

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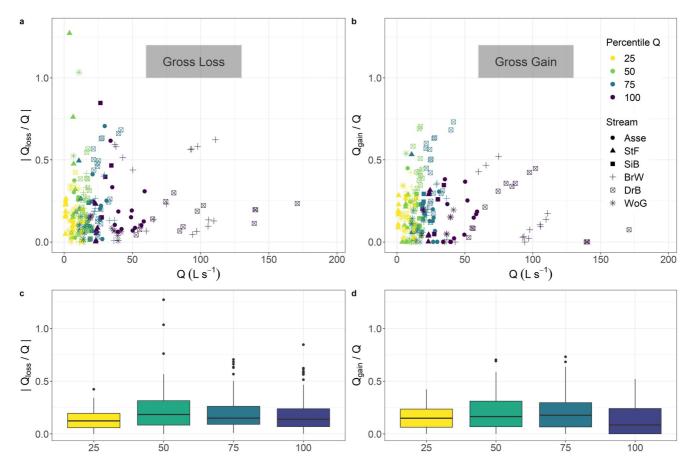


Figure 6. Relative (a) gross loss (absolute values) and (b) gross gain versus discharge, and (c and d) respective percentiles of streamflow. The color coding stands for the quarterly percentiles of discharge. Different shapes correspond to the individual streams.

is still intact and stable, patterns in hydrologic turnover emerge at the reach scale that tend to be persistent throughout the year.

# 4.3. Temporal Variability of Gross Exchange With Flow Regime

Baseflow conditions are considered to facilitate favorable conditions for biogeochemical reactions as in-stream residence times are longer and the fraction of streamflow exchanged with the subsurface is higher than during higher discharge (Bukaveckas, 2007; Covino et al., 2011; Grant et al., 2018). Here, we found that relative gross exchange is highest for moderate discharges (50th–75th percentile, Figures 6c and 6d). Even though the differences are statistically not significant, it is surprising that the relative magnitude of gross gains and losses does not monotonically decrease with increasing discharge (Figures 6a and 6b). This missing decrease contrasts with the relationship between  $Q_{loss}$  and Q found earlier (Covino et al., 2011). Others observed substantial losses for low and intermediate flows (Payn et al., 2009), which also fit our observations. Outliers to this overall picture, that is, occurrence of high exchange with high discharge in some of the agricultural streams (Figures 6a and 6b), might relate to anthropogenic features such as tile drains or fine sediment loadings that could overprint stream morphology and impact water exchange patterns (Nowinski et al., 2011; Schälchli, 1992; Zimmer & McGlynn, 2018).

#### 4.4. Limitations and Uncertainties of the Study

The sensitivity of the dilution gauging method is dominated by (a) experimental error and (b) the given environmental conditions that impact the quality of the signal. (a) Experimental error may be noise in the BTC caused by, for example, turbulent fine sediment elevating the EC background when stepping into the stream or incomplete mixing of the injected tracer solution (e.g., water temperatures <1°C or too short mixing length). (b), especially

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concerns high background EC levels and slightly varying discharge conditions during the experiment causing unstable EC conditions. We assume that especially tracer recovery rates greater than 100% of the injected mass may indicate such problems (in our study 11% rejected from analysis). In the literature, we found estimates of comparable magnitude for error approximation in conservative tracer injection studies (e.g., Schmadel, 2009).

The accuracy of the sensors and, subsequently, detection limits for long BTC tailing owing to signal-to-noise interferences could involve biased results after integration of potentially truncated BTCs (e.g., Drummond et al., 2012; Jawitz, 2004; Schmadel et al., 2016; Ward, Gooseff, et al., 2013). We are aware of the incomplete nature of the BTC signal but found that BTC tailing  $(t_{85}-t_{100})$  in our study did contain a tracer mass representing volumes of 2,86 L s<sup>-1</sup> (mean) and 4,79 L s<sup>-1</sup> (median), respectively (Table S4 in Supporting Information S1). The net change was almost not impacted, and gross losses and gains did not change their behavior, maintaining direction and relative magnitude. In agreement with those results, we used the full integration time  $(t_{100})$  for our analysis. We suggest that an estimate of uncertainty for the mass balance method could be the fraction of data that represented physically impossible cases of gross loss/gain greater/smaller than zero. Roughly 9% of our data account for this.

### 4.5. Implications for Stream Ecology and Nutrient Processing Studies

Methods of nutrient spiraling, which consider a net change in discharge or (measurable) groundwater contributions in their calculations, yet represent appreciatory simple and easy to perform techniques (Graeber et al., 2018; Martí et al., 2004; Roberts & Mulholland, 2007; Runkel, 2007; von Schiller et al., 2011). However, the underlying assumption of parity in net and gross exchange is not generally justified. Direct comparison of gross and net water exchange highlights the unperceived yet actual stream water exchange when using net-hydrological approaches. In over 90% of our data, median gross exchange was five times larger than median net exchange. In addition, we observed that the highest relative exchange coincided with intermediate values of streamflow, which occur mainly during spring and autumn. Those are also times of high biogeochemical activity (e.g., spring and late summer blooms of phytoplankton, higher bacterial activity with increased temperatures, e.g., Bott, 1975). If both hydrological exchange and biogeochemical processes are high, unraveling their contributions to apparent nutrient uptake becomes even more critical for interpreting the integral signal of in-stream nutrient processing. Depending on the composition of the gaining water, a dilution or enrichment of the nutrient concentration could occur over distances of a few hundred meters. Such dilution or enrichment would mimic the effects of biological turnover and result in false deductions by nutrient net-uptake calculations. Therefore, we would explicitly incorporate the channel water balance and hydrologic turnover into net nutrient uptake calculations. The quantified gaining and losing fluxes would enable to distinguish between hydrological and biogeochemical contributions to any measured apparent in-stream solute signal. Considering gross exchange will help to better understand the role of stream ecosystems in cycling nutrients and carbon (e.g., von Schiller et al., 2011; Graeber et al., 2018) and further biologically reactive solutes, such as pesticides (e.g., Rasmussen et al., 2012).

From other studies, we know that stream-groundwater exchange may be locally focused (Conant., 2004; Fleckenstein et al., 2006; Kalbus et al., 2009; Schmidt et al., 2006). Zhang et al. (2021) could evaluate the interplay of stream-groundwater exchange and in-stream solute loadings with their model for an extended version of hydrologic turnover. They obtained ratios of 20/80 for contributing reach length to catchment outlet water composition. Hence, concomitant high gross gains and elevated groundwater nutrient concentrations could entail high in-stream nutrient loadings even if this happened over short distances. In combination with anthropogenic features, such as tile drains, the biogeochemical signal generated by in-stream processing might be additionally overprinted. In conclusion, the interplay of different anthropogenic impacts in combination with (natural) geomorphological characteristics of the stream make up a background noise that may impede a straightforward interpretation of nutrient net-uptake and we urge for the application of combined approaches.

# 5. Conclusions

This study intended to highlight the importance of hydrologic turnover at the reach scale. For the majority of our data, gross gains and losses were substantially higher than net exchange. The first-order streams in this study were preferentially net-losing and had hydrologic turnover distances below one km. In the forested streams, we found higher turnover and spatiotemporally persistent patterns in gross exchange, characterized by coarse-grained

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streambed sediments and stream morphological features such as pools and riffles. The agricultural streams revealed an opposite pattern, as reaches with the highest magnitudes of gross exchange varied over time, not showing a consistent spatial pattern. Surprisingly, the highest relative gross gains and losses for all streams occurred at moderate streamflow in spring and autumn and did not monotonically decrease with discharge as previous studies suggest.

Our results contribute to the understanding of hydrological processes at the reach scale and their implications for solute processing. A detailed evaluation of gross losses and gains is inevitable to avoid misinterpretation of results for nutrient net-uptake, providing the potential to distinguish between hydrological and biogeochemical contributions to the measured apparent in-stream solute signal.

# **Conflict of Interest**

The authors declare no conflicts of interest relevant to this study.

# **Data Availability Statement**

The underlying datasets for this research are available in Jähkel (2021).

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