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Interactions between suspended kaolinite deposition and hyporheic exchange flux under losing and gaining flow conditions

Aryeh Fox[1], Aaron I. Packman[2], Fulvio Boano[3], Colin B. Phillips[2], & Shai Arnon[1]

[1] Zuckerberg Institute for Water Research, The J. Blaustein Institutes for Desert Research, Ben-Gurion University of the Negev, Israel

[2] Department of Civil and Environmental Engineering, Northwestern University, Evanston, IL 60208, USA

[3]Department of Environment, Land and Infrastructure Engineering, Politecnico di Torino, Turin, Italy

Corresponding author: Shai Arnon (sarnon@bgu.ac.il); Aryeh Fox (aryehfoxxx@gmail.com)

Key Points:

- Suspended particle deposition dynamics were different under losing flow conditions as compared to gaining flow conditions.
- Pore clogging processes significantly reduce the hyporheic exchange flux for all tested flow conditions.
- Experiments reveal that fine particle deposition and clogging causes increased subsurface lateral flow.

Abstract

Fine particle deposition and streambed clogging affect many ecological and biogeochemical processes, but little is known about the effects of groundwater flow into and out of rivers on clogging. We evaluated the effects of losing and gaining flow on the deposition of suspended kaolinite clay particles in a sand streambed, and the resulting changes in rates and patterns of hyporheic exchange flux (HEF). Observations of clay deposition from the water column, clay accumulation in the streambed sediments, and water exchange with the bed demonstrated that clay deposition in the bed substantially reduced both HEF and the size of the hyporheic zone. Clay deposition and HEF were strongly coupled, leading to rapid clogging in areas of water and clay influx into the bed. Local clogging diverted exchanged water laterally, producing clay deposit layers that reduced vertical hyporheic flow and favored horizontal flow. Under gaining conditions, HEF was spatially constrained by upwelling water, which focused clay deposition in a small region on the upstream side of each bed form. Because the area of inflow into the bed was smallest under gaining conditions, local clogging required less clay mass under gaining conditions than neutral or losing conditions. These results indicate that losing and gaining flow conditions need to be considered in assessments of hyporheic exchange, fine particle dynamics in streams, and streambed clogging and restoration.

1. Introduction

Suspended sediment is constantly moving in streams and rivers and is a vital part of their aquatic ecosystems (Brunke, 1999). While all types and sizes of sediment can be transported by rivers, suspended fine particles (< 10 μm), which are often comprised of clay particles or organic matter, are ubiquitously found throughout river networks even under low flow conditions. Land use changes due to human activity have drastically increased the amount of fine particles that are traveling through streams and rivers (Wohl, 2015). Under certain conditions, fine particles can accumulate in streambeds within coarser material, a process that is commonly termed siltation or clogging (e.g., Brunke, 1999; Mathers et al., 2014; Wharton et al., 2017). Streambed clogging reduces the hydraulic conductivity (K_s) of the streambed affecting a myriad of processes including: water fluxes into and out of the channel bed and banks (hyporheic exchange) (Findlay, 1995; Rehg et al., 2005), the hydraulic connections between streams and groundwater (Veličković, 2005), biogeochemical processes (Mendoza-lera & Datry, 2017; Mendoza-Lera & Mutz, 2013; Navel et al., 2011; Nogaro et al., 2010), and a wide variety of ecological processes (Boulton et al., 2010; Mathers et al., 2014).

Suspended fine particle deposition in streams is controlled by stream flow conditions and sediment characteristics (Hünken & Mutz, 2007; Packman et al., 2000). For example, slow stream flow conditions favors particle deposition due to settling (García, 2008). For increasing flow velocities, advective particle transport and deposition become increasingly important since hyporheic exchange flux (HEF) increases exponentially with streamflow velocity (Arnon et al., 2013; Packman et al., 2004). The effect of overlying water velocity on fine particle deposition has been extensively studied (e.g., Fries & Trowbridge, 2003; Rehg et al., 2005; Stewardson et al., 2016). Recently, it has been suggested that the exchange of water between the stream and the groundwater (i.e., losing or gaining flow conditions) also plays a significant role in fine particle depositional processes (Chen et al., 2013; Partington et al., 2017), hyporheic exchange, and biogeochemical processes (Azizian et al., 2017; Cardenas & Wilson, 2007; De Falco et al., 2016; Fox et al., 2014; Trauth & Fleckenstein, 2017). Some field surveys have demonstrated that the K_s of the stream bed is lower under losing conditions, but the mechanisms controlling this process are not known (Chen et al., 2013; Dong et al., 2012; Simpson & Meixner, 2012).

The aforementioned studies provide evidence for the influence of stream-groundwater interactions on streambed clogging. However, because the history of flow conditions and streambed characteristics are generally not known in field studies, they do not provide an unambiguous explanation of the governing processes. In order to fill this gap, we conducted controlled flume experiments to quantify how losing and gaining flow conditions affect the deposition of suspended clay, and how this particle deposition influences HEF. We postulated that particle deposition should increase with increasing HEF, but streambed clogging depends on local deposition patterns controlled by interactions between gaining/losing fluxes and bed form-induced HEF.

2. Materials and Methods

2.1 Experimental set up

Interactions between kaolinite particle deposition and HEF were studied in a 640 cm long and 29 cm wide recirculating flume (Supporting Information I, Fox et al., 2016). The flume was filled with natural silica sand (384 μm mean diameter) to form a 20-cm deep streambed over a 540-cm of the flume channel. The bed surface was manually formed into dune-shaped bed forms, which were 15 cm long and 1.5 cm tall with the crest positioned 10 cm from the downstream trough. The porosity of the sand was 0.33 and the K_s is 0.12 cm s^{-1}. The sand used in all experiments was washed with a weak acid and base solution in order to remove residual salts, similar to the procedures that were described by Packman et al. (1997). Average water depth measured from the water surface to the bed form crest was 9 cm. Water in the flume was recirculated using a centrifugal pump (Lowara CEA 370/2/A) and discharge was measured with a magnetic flow meter within the return pipe (Siemens SITRANS F mag 5000). To enforce losing or gaining flow conditions in the streambed, a drainage system was constructed on the bottom of the flume and connected to a peristaltic pump, which enables control of the direction and magnitude of vertical flow through the streambed (i.e., losing and gaining flux)(Fox et al., 2014). The volume of water in the flume was maintained constant by compensating for gains or losses by pumping water into or out of the main channel with an additional peristaltic pump for losing and gaining conditions, respectively (Supporting Information I). The bed form dimensions mentioned above, as well as the flow conditions used in this study, are typical of sand-bed streams (e.g., Harvey et al., 2012; Hünken & Mutz, 2007; Mutz, 2003; Mutz, 2000; Stofleth et al., 2008; Strommer & Smock, 1989; Wörman et al., 2007, and references within).

2.2. Experimental approach

Three sets of flume experiments were conducted with an average overlying water velocity of 15 cm s^{-1} (calculated by dividing the discharge by the channel cross-section area that was measured at the bed form crest). One set of experiments was conducted under losing flux (q_L) of 12.5 cm day^{-1}, another under gaining flux (q_G) of 12.5 cm day^{-1}, and the third conducted under neutral conditions (i.e., without imposing a vertical flux). q_L and q_G were calculated by dividing the imposed vertical discharge by the streambed surface area. A detailed description of the experiments, including a detailed time line, experimental procedures, and preparations before each tracer experiment are given in the Supporting Information II. Briefly, in each set of experiments, the flow conditions were set and an initial characterization was conducted by measuring HEF with a salt tracer, and by visualizing the flow patterns in the streambed using a dye tracer (see details in section 2.3). After the initial characterization, consecutive additions of suspended clay particles (kaolinite) were performed until HEF substantially reduced. The extent of clay deposition was recorded continuously by measuring water turbidity. HEF was measured after each addition of kaolinite. Dye injections were used to visualize HEF both before and after each set of kaolinite additions. Finally, streambed core samples were collected along the bed form to evaluate the spatial distribution of kaolinite deposits.

2.3. Particle, salt and dye tracer additions

Kaolinite deposition rates were measured by adding kaolinite to the surface water and measuring the decline in concentration over time. Each individual addition contained 80 g of kaolinite (cat. 470025-474, Ward's Natural Science, USA) suspended in five L of deionized water containing 10 mM NaCl. The suspension was vigorously mixed for 24 hours prior to the experiment, and then added into the endwell over the duration of a single water recirculation time in the flume to ensure efficient mixing of kaolinite in the surface water. After dilution in the flume, the background electrolyte concentration was approximately 3 mM NaCl, which is far below the critical coagulation concentration (CCC) of aqueous suspensions of kaolinite (Tombácz & Szekeres, 2006). Kaolinite concentrations in the surface water were measured continuously (every 30 seconds) by a turbidity sensor (TurboVis, Xylem, UK), which was calibrated with known concentrations of kaolinite samples prior to the experiments.

Salt tracer additions were performed to measure HEF. Each tracer solution contained 120 gr of NaCl dissolved in 5 L of deionized water, which was added to the flume similarly to the kaolinite solution. The concentration of the salt in the water was monitored with an EC meter (multi 3430 logger, WTW, UK). The EC in these experiments was maintained between 300-1000 $\mu S\ cm^{-1}$.

Dye additions were used in order to visualize the exchange flow paths before and after kaolinite deposition in the streambed. 25 g of Brilliant Blue dye was dissolved in 5 L of water and added to the flume. The dye penetration into the sediment was recorded for 24 hours by sequential photographs taken every 30 s through the glass side walls of the flume.

2.4. Distribution of kaolinite in the streambed

We assessed the distribution of kaolinite in the streambed by taking core samples along each bed form after each experiment. Triplicate samples were taken from four sections along the bed form (within 0-3, 4-7, 8-11, and 12-15 cm from trough to trough, Supporting Information III). For orientation, location 0 cm is the trough, 4 cm is on the stoss side, 8 cm is close to the crest, and 12 cm is on the lee side of the bed form. Modified plastic syringes with a diameter of 2.9 cm and a length of 10 cm were used to collect core samples (Supporting Information III). Before taking cores, the flow was stopped and the water level was gently lowered in order not to disturb the surface layer of the streambed. Syringes were then inserted into the bed, sealed from the bottom and then carefully removed in order not to disturb the structure of the core sample. The cores were then sectioned every 0.5 cm, which yielded samples of approximately 7 g of wet sand. Extraction of kaolinite from the sand was done by vigorously mixing each section with 50 mL of deionized water. The concentration of kaolinite in the water was measured with a spectrophotometer (Evolution 220, Thermo Scientific, USA) by calibrating kaolinite concentrations to absorbance at 600 nm.

2.5. Data analysis

HEF was quantified using mass balance equations based on the theory presented by Elliott and Brooks (1997) and extended by Fox et al. (2014). The latter developed a method that separates the effect of the imposed losing/gaining flux from the HEF. Images from all the dye additions were analyzed for the dye distribution in the streambed, using a MATLAB batch image analysis routine developed by Fox et al. (2016). Comparing time-lapse images enabled us to follow the spatial and temporal changes in HEF before and after kaolinite deposition. The extent of the hyporheic zone was calculated as the area of dye penetration in the bed observed 24 hours, after dye injections.

We visualized the initial porewater velocity field using a numerical model developed previously for HEF under gaining and losing conditions (Boano et al., 2018). The model was built in COMSOL to reproduce 2D laminar water flow below a periodic bed form. Following previous studies (e.g., Elliott & Brooks, 1997), a sinusoidal function was used to describe the hydraulic head distribution along the bed form profile, and a constant flux boundary condition was set at the domain bottom to match the flux imposed in each laboratory experiment. A complete description of the model can be found in the Supporting Information (Section IV).

Kaolinite flux into the bed was calculated as the average removal of clay mass from the surface water per time normalized by the streambed surface area. The effect of kaolinite deposition on HEFs under gaining, neutral and losing conditions was evaluated by fitting a linear function to the reduction of HEF over time. Differences in HEF between flow conditions were evaluated by comparing the sum-of-squares for each independent fit, and the combined fit that was calculated using the extra sum-of-squares F test in the statistical software GraphPad Prism (version 5).

3 Results and Discussion

Dye propagation in the clean sand before the kaolinite additions was observed with time lapse photography to assess the evolution of porewater flow patterns (Figure 1a-c). Dye fronts propagated quickly into the bed for the first few hours. The dye propagation rate decreased quickly under gaining conditions, and ceased completely when the front reached the bottom of the bed under losing conditions, but continued under neutral conditions even after 24 hours (Figure 1a-c and Supporting Information V). Under gaining and neutral conditions, the dye distribution within the bed is solely related to HEF, since the upwelling water was dye-free. Thus, the photographs show nicely how upwelling flow under gaining conditions suppressed the size of the hyporheic zone as compared to neutral conditions. Under losing conditions, the dye patterns reflect a combination of HEF and the imposed losing flux, which prevents determination of the extent of the hyporheic zone using this method.

Comparing the dye images with modeled flow fields reveals a good match under gaining and neutral flow conditions, which is illustrated by the similar size and shapes of the hyporheic zone (Figure 1a, b, d, e). The flow fields show that water infiltrated into the subsurface on the stoss side of the bed forms and returned back to the stream in the lee side, but also on the lower parts of the stoss side due to some backwards flow paths (lower left side of the images in Figure 1 d-e, and also Cardenas & Wilson, 2007). Under gaining flow conditions, upwelling groundwater enclosed the hyporheic zone from both sides, and entered the stream mostly within the lee side. Under losing conditions, downwelling flow entered the bed on the entire stoss side, but bed form-induced hyporheic exchange still occurred with flow paths returning to the stream on the lee side (Figure 1f).

Differences in the observed dye propagation rates before and after kaolinite addition clearly demonstrate that clogging affected the subsurface flow (Figure 1a-c and Supporting Information V). Dye fronts propagated in the bed more slowly after the kaolinite additions under all flow conditions, indicating that clay deposition decreased HEF and the size of the hyporheic zone. After the kaolinite addition, the extent of dye penetration determined from images (Figure 1 a-c) was 3%, 13%, and 75% of the bed after 480 minutes under gaining, neutral, and losing flow conditions, respectively. The same extent of penetration in the clean sand bed (before clay addition) required only 20 minutes for gaining conditions, 60 minutes for neutral conditions and 460 minutes for losing conditions (Supporting Information VI). The extent of dye penetration after 24 hours of kaolinite deposition was markedly smaller for the gaining and neutral conditions than the losing conditions (Supporting Information V).

Decreases in HEF coincided with decreases in kaolinite deposition flux (Figure 2). In each kaolinite addition, the kaolinite concentration in the surface water column declined rapidly for the first few hours, and then the deposition rate decayed slowly for the remainder of each experiment. In all cases, the clay concentration in the water column decreased by at least 50% after 25 hours (Figure 2a). The largest amount of kaolinite deposition occurred under losing conditions, and the smallest occurred under gaining conditions (Supporting Information VII). Kaolinite fluxes into the bed averaged 3.25, 3.3, and 4.12 g $m^{-2}hr^{-1}$ over the first five hours under gaining, neutral and losing conditions, respectively, and thereafter decreased to 0.49, 0.76, and 0.82 g $m^{-2}hr^{-1}$, respectively, over the remaining 19 hours of each clay addition. These differences in kaolinite deposition rates under neutral, losing, and gaining flow conditions reflect the total water flux into the bed for each condition. Under neutral conditions, the exchange flux is purely driven by the bed structure and the overlying flow conditions (Elliott & Brooks, 1997; Fox et al., 2014; Packman & Salehin, 2003). Here, the neutral bed form-induced HEF was 17 cm d^{-1}. Losing/gaining fluxes are superimposed on the bed form-induced hyporheic exchange, which reduces HEF and redistributes the porewater flow field (Figure 1 d-f, Cardenas & Wilson, 2007; Fox et al., 2016; Trauth et al., 2013). Under gaining and neutral flow conditions, the total flux into the bed equals the measured HEF, while under losing conditions it is the sum of HEF and the losing flux. Here, the imposed losing and gaining fluxes were both 12.5 cm d^{-1}. Therefore, the total water fluxes from the surface water into the clean sand bed (before kaolinite addition) were smallest under gaining conditions (12 cm d^{-1}), intermediate under neutral conditions (17 cm d^{-1}), and greatest under losing conditions (24.5 cm d^{-1}). The trends in these imposed total water fluxes into the beds follow observed trends in kaolinite deposition in the bed. Finally, consecutive additions of the same kaolinite mass resulted in a linear reduction in HEF under all flow conditions, with a greater rate of decrease under gaining conditions than under neutral and losing conditions (Figure 2b).

These observations can be understood as the coupling between hyporheic exchange and particle deposition. Advective flux into the bed carries suspended kaolinite particles that become filtered along hyporheic flow paths (Elimelech, 1998; Packman et al., 2000). This filtration ultimately results in deposition and clogging in regions of water inflow to the bed, which decreases HEF and porewater flow (Packman and MacKay, 2003). While the clay accumulation was very small, yielding average clay mass fractions in the total bed of 0.036%, 0.041%, and 0.052% under gaining, neutral and losing conditions, respectively, accumulation was locally much greater in regions of porewater inflow into the bed. Streambed core samples showed that kaolinite deposition was more pronounced along the surface of the stoss side of the bed forms under all flow conditions (Figure 1g-i and supporting Information VIII). Flow simulations indicate that this region is where most of the advective hyporheic flux enters the bed (Figure 1d-f). Gaining conditions yielded clay concentrations of >1% mass fraction of bed sediments in a small region in the area of greatest HEF: the middle of the stoss side of bed forms (Figure 1g). Under neutral conditions, concentrations of kaolinite also ranged between 1.0-1.2%, but occupied a much larger area covering the majority of the stoss side of the bed form, with lower amounts of deposition in the region where flow leaves the bed (Fig. 1h). Kaolinite deposition under losing conditions was also focused along hyporheic flow paths (Figure 1f), but occurred over a wider region of the streambed surface and yielded clay accumulation deeper in the bed. Under losing conditions, kaolinite concentrations ranged between 1.2-1.5% along the stoss side, with lower concentrations observed along the lee side and at the crest (Figure 1i).

The horizontal layered structure of kaolinite deposits not only decreased HEF and vertical dye penetration, but also led to an increase in the lateral spreading of exchanged dye. Under neutral conditions, regions of hyporheic upwelling flow paths, shifted from the recirculation zone on the lee side of each bed form – the location predicted for homogeneous streambeds – to underneath the crest (indicated by dye-free regions in Figure 1b). Under losing conditions, the dye-free upwelling zones between bed forms disappeared as a result of clogging (Figure 1c). While these upwelling zones were visible after four hours of exchange in the clean bed, they disappeared in the clogged bed after approximately one hour (Figure 1c). These shifts in porewater flow patterns reflect an evolution of the hyporheic exchange flow trajectories due to the clogging of pore spaces. In particular, increased horizontal spreading of the dye fronts indicates that clay deposition in regions of porewater inflow reduced HEF, decreased vertical dye penetration, and shifted hyporheic flow horizontally. Counterintuitively, HEF decreased more quickly under gaining conditions and required less clay mass than neutral or losing conditions, despite the fact that the initial water exchange and clay deposition flux were lowest under gaining conditions (Fig. 2b). This is because the area of hyporheic exchange is much smaller under gaining conditions, and highly constrained by the upflowing water. Under neutral and losing conditions, clay deposition and clogging in the area of water inflow to the bed diverted inflowing water laterally, shifting inflow to other areas of the bed form (Figure 1 b-c) and ultimately producing layered deposits over much of the stoss side of the bed form (Figure 1 h-i). However, under gaining conditions, the region of inflow and deposition is highly limited to a narrow region in the middle of the stoss side of the bed form (Figure 1a) and clay deposition is focused specifically in this region (Figure 1 g). This constraint imposed by upwelling means that clogging of the small region of influx to the bed more readily reduces HEF under gaining conditions than under neutral or losing conditions.

The deposition of fine particles near the streambed surface found here has been commonly observed in both laboratory and field studies (Arnon et al., 2010; Drummond et al., 2014, 2017; Stewardson et al., 2016). An implication of this is that fine particle accumulation within streambeds is highly sensitive to flow events capable of scouring the upper layer of the streambed and remobilizing deposited fine particles. However, particle dynamics during floods remain complex. Fine particles near the surface are often remobilized due to bed mobility and scour, while some particles can be propagated deeper into the streambed where retention times are significantly longer (Drummond et al., 2014). Other studies that have followed the temporal dynamics of streambed K_s have found that streams with less frequent bed disturbances have lower streambed K_s and reduced HEF (Blaschke et al., 2003; Datry et al., 2015; Stewardson et al., 2016). Blaschke et al. (2003) specifically observed in the Danube River that clogging occurred mostly in the upper few centimeters. The deposition patterns observed in our experiments show that this behavior may be caused by the strong deposition and clogging in bed forms under neutral and gaining conditions, and more distributed particle deposition under losing conditions.

The layered depositional structures observed here are also common in stream beds and can be formed by various mechanisms, mostly by depositional patterns during mobile bed conditions (Huggenberger et al., 1998; Powell, 1998). Layered depositional structures in streambeds produce anisotropy that decreases vertical hyporheic exchange and favors flow parallel to deposit layers (Fox et al., 2016; Gomez-Velez et al., 2014; Jesus et al., 2014; Salehin et al., 2004; Zlotnik et al., 2011). Our results indicate that significant anisotropy can develop in immobile beds under constant flow conditions due to the combination of advective exchange and filtration of fine particles, and this will restrict vertical HEF and favor shallow HEF in horizontal layers under bed forms.

These spatial patterns of HEF and clay deposition have several important implications. The impact of clay accumulation on HEF is expected to influence water budgets in streams and connectivity between streams, floodplains, and the underlying aquifers (e.g., Nowinski et al., 2011). Such a reduction in connectivity may negatively affect bank filtration (Goldschneider et al., 2007). The reduced HEF and altered flow patterns will change the residence time of solutes in the streambed, thereby influencing biogeochemical processes. Clogging can result in shallower flow paths and a shrinking of the hyporheic zone (Figure 1), which would induce a thinner oxic zone (Caruso et al., 2017; De Falco et al., 2016; Kaufman et al., 2017). Changes in HEF, flow patterns and chemistry will initiate a response in the ecological communities and their functions. For example, clogging of the hyporheic zone degrades habitat for benthic fauna, which reduces diversity and can affect metabolism and the productivity of the lotic ecosystem (Brunke, 1999; Jones et al., 2015; Mathers et al., 2014). These processes could broadly influence stream ecosystem functions and resilience with implications for management (Nogaro et al., 2010; Wharton et al., 2017). The presence of unclogged sand beds indicates that there must be either very little input of fines to the system or relatively frequent bed sediment transport to resuspend deposited fines. In addition, biological processes, such as bioturbation, can also remobilize fine material and maintain K_s (Nogaro et al., 2006; Song et al., 2010).

Beyond the clogging of sand beds observed here, interactions between HEF and gaining/losing are expected to play a significant role in depositional patterns in coarse high-permeability streambeds, but more fine particle accumulation will be needed to induce clogging. However, fine particles can propagate more deeply into coarse gravel/cobble beds where the probability of remobilization is lower, and coarse beds are also less susceptible to scouring by floods, providing longer periods for fine particle accumulation (Chen et al., 2013; Nowinski et al., 2011). Prior work shows that clogging is important in gravel-bed rivers, but the dynamics of these processes are not well understood because of the difficulty of obtaining *in situ* information on particle deposition and clogging/unclogging processes over the range of scales that are important (Chen et al., 2013; Descloux et al., 2010; Dong et al., 2012). This is because the relevant scales are generally much larger in gravel-bed streams than in sand-bed streams.

This study provides clear evidence that stream-groundwater interactions and deposition of suspended particles are generally coupled over a wide range of scales. Prior studies have shown that fine suspended particles are transported into sandy streambeds, leading to clogging of the streambed in locations of hyporheic inflow (Packman & Mackay, 2003). Here we showed unique experimental evidence that both clay accumulation in the streambed and resulting changes in hyporheic exchange flows strongly interact with larger-scale gaining and losing flows. Kaolinite deposition led to a decrease in HEF, changed patterns of hyporheic exchange, and reduced the size of the hyporheic zone. Following clay deposition, hyporheic flow spread laterally within the near-subsurface region, and propagated outside of the zone of hyporheic exchange identified in the clean bed (prior to clay addition). This lateral hyporheic spreading was observed under all flow conditions, but was more prevalent under gaining and neutral flow conditions.

A major outcome of these observations is that it is essential to evaluate the spatial distribution of K_s when quantifying exchange fluxes and biogeochemical processes within a reach. The spatial distributions of clay in coarse sediment beds are also important for assessing the potential for resuspension when assessing the effects of siltation on hyporheic ecosystems. Including the effects of losing and gaining flow is necessary to ensure that stream restoration will produce the desired outcomes for hyporheic exchange and ecological function. While geomorphic field studies often characterize the bulk fine fraction in streambed sediments, our results show that the clogging process is much more local to the streambed surface, particularly in areas of hyporheic exchange flux into sand beds. Furthermore, the extent of heterogeneity in clogging strongly depends on the pattern of both hyporheic exchange induced by local streambed features (e.g., bed forms) and larger patterns of river gaining and losing. Field studies characterizing the K_s of river reaches for the purpose of assessing clogging and/or restoring degraded hyporheic zones should design sampling schemes to capture the multiscale exchange and clogging behavior shown here. In particular, a greater number of samples will be needed to characterize streambed K_s and clogging in reaches with strong internal geomorphic complexity than in reaches where fine particle deposition and clogging are dominated by general downwelling conditions. Disconnection of near-surface and deeper porewater by formation of clogging deposits that lead to horizontal preferential HEF should also be considered in field site assessments, as these types of flows may be missed by methods that assume homogeneity and/or isotropy, such as estimations of exchange fluxes from measurements of vertical hydraulic gradients in streambeds.

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Figure captions:

Figure 1: Time series photographs of dye penetration into the bed under gaining (a), neutral (b) and losing (c) flow conditions before and after kaolinite additions (end of experiments). Flow was from left to right at an overlying water velocity of 15 cm s^{-1} and the losing/gaining flux was 12.5 cm d^{-1}. The velocity field within the sand before the kaolinite additions is shown below the relevant images (d-f), while the patterns of kaolinite concentrations within the sand at the end of the experiments are shown in images (g-i). Kaolinite concentrations are represented as averages of nine samples (Supporting Information III), while standard deviations of kaolinite concentrations were lower than ±0.38% (Supporting Information VIII).

Figure 2: Reduction in the relative kaolinite concentrations in the surface water during three separate additions (a) and the influence of kaolinite deposition on HEF (b). Each data point in panel b represents one addition of kaolinite while the sand was initially particle-free. The HEF decreases linearly with increasing amounts of kaolinite deposition. All fits had $R^2 > 0.99$ and were statistically different from each other ($p < 0.05$).