J Soils Sediments (2015) 15:2174–2188
DOI 10.1007/s11368-015-1203-6

ADVANCES IN SEDIMENT FINGERPRINTING

# The role of gravel channel beds on the particle size and organic matter selectivity of transported fine-grained sediment: implications for sediment fingerprinting and biogeochemical flux research

**Alexander J. Koiter**[1] **· Philip N. Owens**[2] **· Ellen L. Petticrew**[3] **· David A. Lobb**[4]

Received: 16 May 2015 / Accepted: 12 July 2015 / Published online: 9 August 2015


**Abstract**
*Purpose* The preferential erosion, delivery, and transport of sediment within a drainage basin can result in downstream changes in both particle size and organic matter content. The physical and biological properties of transported and deposited sediment are important considerations in many sediment management and investigative tools, including sediment fingerprinting, and aid in the interpretation of sediment-associated nutrient and contaminant data.
*Materials and methods* A recirculating flume (2×2×40 m) was used to assess the changes in particle size, organic matter content, and geochemical composition of fine-grained sediment (<125 μm) over a 31-h period (representing a travel distance of ~24 km) under three contrasting channel bed conditions. The three channel gravel bed conditions investigated were 0-, 5-, and 40-cm gravel bed depths. Suspended sediment samples were collected throughout the duration of the experiment and gravel-stored sediment were collected along the length of the flume at the end of the experiment. Both suspended and gravel-stored sediment were analyzed for particle size and organic matter content. In addition, suspended sediment samples were analyzed for a broad suite of geochemical elements.
*Results and discussion* The channel bed characteristics had a significant effect on both the particle size and organic matter selectivity of the transported suspended sediment. Furthermore, it was shown that a smooth planar channel bed as compared to a planar gravel bed (i.e., 0- vs. 5-cm gravel treatments) introduced small-scale roughness which resulted in the preferential deposition of larger particles into the channel bed. Increasing the gravel bed from 5 to 40 cm increased the amount of intra-gravel flow and reduced the potential for resuspension resulting in a further reduction in the particle size as well as resulting in a significant increase in the organic matter content of the suspended sediment. The relation between geochemical concentrations and particle size of the suspended sediment, in terms of linearity, magnitude, and direction, were not consistent between the different elements investigated.
*Conclusions* This research helps to understand the processes that control the particle size and organic matter selectivity of fluvial transported sediment. This information is an important part of many sediment management tools, including sediment fingerprinting, as it provides context when selecting sampling sites and interpreting the data they provide. The inconsistent relation between particle size and the concentration of different geochemical elements highlights the uncertainty associated with commonly used particle size correction factors.

**Keywords** Fluvial transport · Sediment geochemistry · Gravel bed · Organic matter · Particle size

Responsible editor: Hugh Smith

✉ Alexander J. Koiter
koiter@unbc.ca

[1] Natural Resources and Environmental Studies Program, University of Northern British Columbia, 3333 University Way, Prince George, BC V2N 4Z9, Canada

[2] Environmental Science Program and Quesnel River Research Centre, University of Northern British Columbia, 3333 University Way, Prince George, BC V2N 4Z9, Canada

[3] Geography Program and Quesnel River Research Centre, University of Northern British Columbia, 3333 University Way, Prince George, BC V2N 4Z9, Canada

[4] Department of Soil Science and Watershed Systems Research Program, University of Manitoba, 13 Freedman Crescent, Winnipeg, MB R3T 2N2, Canada

## 1 Introduction

### 1.1 Sediment management tools and approaches

Particle size and organic matter selectivity, in the context of sediment transport, refers to the preferential detachment, entrainment, transfer, and deposition of sediment based on particle size and organic matter content. Along the sediment cascade, sediment mobilization on the hillslopes, storage within the riparian zones, and the fluvial transport in downstream rivers are all selective processes (Slattery and Burt 1997; Koiter et al. 2013a). Information on the processes of particle size and organic matter selectivity is needed for watershed management tools including sediment budgets, geomorphic connectivity, and sediment fingerprinting (Stone and Walling 1997; Di Stefano and Ferro 2002; Fryirs 2013). Many of the watershed tools commonly used can be considered blackbox concepts because there is little consideration as to the processes that link the inputs to the outputs, and in particular, the process of particle selectivity is often overlooked (Walling 1983; Koiter et al. 2013a).

The sediment source fingerprinting technique uses naturally occurring physical and biogeochemical properties as tracers to discriminate between sediment sources and to subsequently estimate the contribution from each sediment source to sediment collected downstream (see Davis and Fox 2009 for a review of sediment fingerprinting). Ideally, the sediment properties selected as tracers do not change (i.e., exhibit conservative behavior) between the source of sediment and the downstream sediment collection so that a direct comparison can be made. However, particle size and organic matter transport selectivity can result in the non-conservative behavior of sediment properties as selectivity tends to result in downstream fining and an enrichment of organic matter content. Both of these properties have been shown to have an effect on geochemical and radionuclide tracer concentrations within the sediment, and this is primarily due to the increase in specific surface area (SSA) and chemical reactivity (Horowitz 1991). The importance of this phenomenon has been recognized within the sediment source fingerprinting community as correction factors for both particle size and organic matter are commonly used. These correction factors are typically based on the ratio of SSA and organic matter content between collected in-stream sediment and potential sediment sources (e.g., Collins et al. 1997). However, the use of these correction factors are increasingly being criticized as they have not been widely tested and have little basis in watershed processes (Koiter et al. 2013a; Smith and Blake 2014).

The importance of incorporating process-based understanding of the changes in physical and biogeochemical properties of sediment during transport through watersheds has been increasingly recognized as a critical step in linking sediment sources to in-stream sediment in a reliable and robust manner. This special issue, Advances in Sediment Fingerprinting, in the *Journal of Soils and Sediments* is evidence of this as many of the research articles focus on the testing of assumptions that link sources to sediment. For example, Pulley et al. (2015) investigated the use of different groups of tracers (e.g., geochemistry, radiochemistry, mineral magnetics) on the apportionment results; Laceby et al. (2015) compared a statistical and geological approach to the tracer selection process; Sherriff et al. (2015) examined the uncertainty with respect to changes in particle size and organic matter content as well as geochemical transformations on the apportionment results; and Wilkinson et al. (2015) incorporated the erosional history in characterizing sediment source tracer concentrations. The present research compliments the other contributions in this special issue by investigating the particle size and organic matter selectivity of fluvial transported sediment and its subsequent influence on the geochemical concentrations of suspended sediment.

The sediment delivery ratios for watersheds are particle size dependent as smaller particle size classes tend to have higher delivery ratios as the coarser sediment fraction is preferentially deposited. For example, Walling (1983) found that within the Jackmoor Brook watershed (9.8 km$^2$) located in Devon, England, very fine-grained sediments (<1 μm) had a delivery ratio of 100 % while coarser sediment (20–63 μm) had a delivery ratio of only 30 %. Furthermore, the suspended sediment within the Jackmoor Brook watershed was also found to be enriched in organic matter by 60 % as compared to the sources of sediment. Stone and Walling (1997) describe the importance of particle size distribution and selectivity in assessing the delivery and transport patterns of different particle size fractions and its subsequent importance in constructing and interpreting sediment budgets for watersheds. For example, Bainbridge et al. (2014) constructed a suspended sediment budget for three different particle size classes (<4, 4–16, and >16 μm) for the Burdekin River watershed (130,400 km$^2$) located in north-east Queensland, Australia. It was found that a dam, located mid-catchment, trapped approximately 66 % of the total incoming sediment; however, the dam resulted in the preferential trapping of the coarsest sediment size class (92 % trapped) as compared to the finest sediment size class (33 % trapped). This preferential deposition of the coarser sediment resulted in 80 % of the total sediment being exported at the outlet of the watershed being comprised of sediment <16 μm despite coarser sediment inputs from tributaries downstream of the dam. This research highlights the importance of considering watershed characteristics when selecting sampling locations and interpreting the data.

The process of particle size and organic matter selectivity also needs to be considered when assessing data on carbon (C), nutrient, contaminant, and geochemical fluxes at the watershed scale. The fluvial transport of organic and inorganic C are key parts of the global C and nutrient cycles. Approximately $1.9 \times 10^{12}$ kg of C (inorganic/organic and dissolved/particulate forms) from terrestrial sources is delivered to surface waterways on an annual basis (Cole et al. 2007). However, only half of this amount is delivered to the ocean, a further ~40 % is returned to the atmosphere, and the remaining ~10 % is stored as deposited sediment in lakes, reservoirs, and rivers. The predominant form of sediment-stored carbon in freshwater systems is particulate organic C (POC), and lakes represent the largest and the most understood and quantified C storage compartment. In contrast, the role of C storage in rivers is smaller and is less understood and quantified (Cole et al. 2007; Tranvik et al. 2009). The particle size and organic matter content of transported sediment are important components of the overall characterization of the spatial and temporal patterns of C storage as these properties drive the settling velocities of sediment (e.g., Young and Huryn 1997). For example, in a 75-km$^2$ catchment, located in south-eastern Queensland, Australia, Garzon-Garcia et al. (2015) found that there were changes in the C content of the sediments for different flow magnitudes. This indicates that there was particle size and organic matter selectivity occurring which resulted in higher C content sediment being transported during lower flow events as compared to higher flow events.

Sediment-associated nutrients, particularly phosphorus (P), are an important nutrient source in the external and internal loading of lakes and rivers and can result in eutrophication (Withers and Jarvie 2008). The concentration of bioavailable P is correlated with particle size, with smaller particles having higher concentrations of P; this coupled with the preferential transport of fine-grained sediment has important implications for the linking of terrestrial P sources to surface waterways and its management (Kerr et al. 2011). For example, Ockenden et al. (2014) found that both the total nitrogen (N) and P concentrations of sediment trapped in 10 on-farm constructed wetlands were inversely related to median particle size. However, the total mass of accumulated N and P within the wetlands was dependent on the total mass of sediment trapped. Similar to nutrients, many contaminants including metals, organic chemicals, pathogens, and pharmaceuticals also tend to be concentrated in the fine-grained and organic-rich sediment (Oliver et al. 2007; Maskaoui and Zhou 2010; Zheng et al. 2012).

### 1.2 Fluvial transport of fine-grained sediment

Within the fluvial environment, fine-grained sediment (<63 μm) transport and depositional patterns are primarily driven by the settling velocity of sediment particles (Naden 2010). The settling velocity of sediment is mainly influenced by the size of the particles as the smaller particles tend to remain in suspension as the masses of the particles are counteracted by the upward momentum created by eddies and turbulence. The composition, specifically the clay and organic matter content, of sediment is also an important factor controlling the settling velocity as these components can act as binding agents aggregating individual particles into larger aggregate particles. Aggregation can influence the settling velocity though changes in size, density, shape, and internal structure of sediment particles (Droppo et al. 1997).

In addition to particle size, other important aspects of fine-grained sediment hydrodynamics include the velocity and transport capacity of the flow. As there is generally an overall downstream reduction in these two fluvial characteristics, the competence of streams to entrain and transport larger particle is reduced; therefore, a downstream fining/gradation of sediment is often observed as the coarser particles are preferentially deposited upstream (i.e., hydraulic sorting). In addition to hydraulic sorting, abrasion and breakage of sediment particles can also result in the downstream reduction in particle size. This is an important process in watersheds containing low strength material (e.g., shale deposits), and it can be difficult to separate the processes of selectivity from abrasion and breakage (Koiter et al. 2013b). There can also be temporal variation in the selective nature of fluvial transport processes due to increases in precipitation and runoff which result in greater flow velocities and transport capacity (Walling et al. 1992). Similar to changes in the particle size distribution, there is generally an overall downstream increase in the organic matter content of the sediment (Rhoton et al. 2006). The preferential transport of organic matter is largely due to the lower density and the subsequent slower settling velocity of organic-rich particles as compared to mineral particles of a similar size.

Reach-scale features can also have a local influence on fine-grained sediment dynamics. These features include longitudinal (e.g., pool and riffle sequence, change in slope), lateral (e.g., flow characteristics around a meander, changes in channel width), and vertical (e.g., due to channel bed roughness, armored layers) variations in flow velocities. These local variations result in the deposition of fine-grained sediments in features including alluvial fans, channel bars, floodplains, and infiltration into the interstices of gravels. Much of the literature focuses on the infiltration of sand-sized particles into the gravel matrix (e.g., Wooster et al. 2008; Grams and Wilcock 2014) and less on the infiltration of fine-grained sediment (<63 μm) as this material is often considered to move though a river system unimpeded (i.e., wash load). However, fine-grained sediment storage within the gravel bed is an important consideration as it can reduce hyporheic exchange, compromise invertebrate and fish habitat, and act as a legacy source of sediment-associated nutrients and contaminants (Rehg et al. 2005; Rex and Petticrew 2008; Albers and Petticrew 2012).

In this study, the particle size and organic matter selectivity of fluvial transported fine-grained sediment was investigated under three contrasting gravel channel bed conditions using a large recirculating flume. The objectives of this research were to investigate: (1) the changes in the concentration, particle size distribution, and organic matter content of suspended and bed sediment over distance traveled; (2) the relation between particle size and organic matter content of the suspended sediment; (3) the changes in the geochemical composition of the suspended sediment; and (4) the ability of a particle size correction factor to account for the changes in suspended sediment geochemistry.

## 2 Methods

### 2.1 Flume characteristics

A recirculating flume located at the Quesnel River Research Centre (QRRC), Likely, British Columbia (BC), Canada (52°37′08.2″N 121°35′33.3″W) was used to investigate the change in physical and biogeochemical properties of fine-grained sediment over time (distance traveled) under three different channel bed conditions. The recirculating flume was constructed from concrete and had dimensions of 2×2×40 m with a gradient of 0.05 % (see Fig. 1 for a schematic of the flume setup). Water was recirculated using two centrifugal pumps (Goulds, Model 3656 LH 54 BF; ITT Industrial Process, NY, USA) with a discharge of 4500 L $min^{-1}$, and large aluminum screens (1 mm opening) were placed at the front and back of the flume to direct the flow of water and dissipate turbulence caused by the pumping of water. The flumes were filled with water to a depth of 15 to 20 cm (above the channel bed) and had a velocity of 25 to 20 cm $s^{-1}$ (at 60 % of the depth from the surface) measured with a propeller-driven flow meter (Swoffer Instruments Inc., WA, USA) at 9 and 27 m from the front of the flume, respectively. The water was supplied from on-site groundwater wells and had a neutral pH (7.15), low conductivity (180 µs $cm^{-1}$), and was devoid of any measurable particulate matter.

The gravel used for the channel bed material was sourced from a local aggregate quarry, and the shape was characterized by selecting 100 stones at random and measuring the three axes of each stone. The gravel were determined to be unimodal and spherical to slightly discoid in shape using the Zingg classification (Zingg 1935) with a mean diameter (intermediate axis; $D_{50}$) of 3.2±0.7 cm (±1 SD). The porosity of the channel bed gravel was 30 % and was measured using the water displacement method (12-L gravel sample). The gravel was washed prior to placement in the flume to remove any fine-grained sediment; also, the gravel and the flume were cleaned in between experimental runs. The lack of fine-grained sediment within the gravel may represent conditions after flushing flows or recent movement of the channel bed. The three channel bed treatments investigated were (1) 0-cm gravel bed (0 $D_{50}$), (2) 5-cm gravel bed (1.5 $D_{50}$), and (3) 40-cm gravel bed (12.5 $D_{50}$) which can be considered as representing end-members in terms of channel bed porosity and sediment storage capacity. This allowed for the examination of bed surface roughness, intra-gravel flows, and sediment storage capacity on particle selectivity. The gravel depth was uniform throughout the length of the flume creating a flat/planar bed form.

### 2.2 Sediment addition description

The fine-grained sediment that was added to the flume were soil surface scrapes (0–5 cm) collected in the lower slope position at the edge of an agricultural field next to the riparian area, in the Beaver Valley near Likely, BC (52°30′27.8″N 121°52′14.0″W), to represent sediment likely to be eroded and delivered to surface waterways. The soil was air-dried and passed through a 2-mm sieve to remove stones and vegetation (e.g., grasses). The air-dried soil (200 g aliquots) was slaked in water for 10 min prior to being placed in a 125-µm stainless steel sieve and raised and lowered by hand in a vertical motion with an approximate displacement of 10 cm in a container with 15 L of water for 10 min to obtain water-stable aggregates <125 µm in size (methods based on Angers et al. 2007). For each treatment, a total of 6.5 kg of air-dried soil

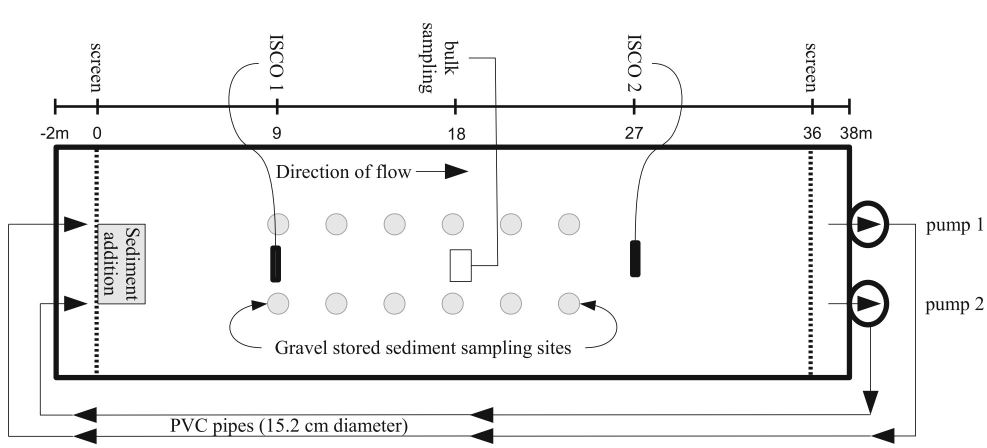


**Fig. 1** Recirculating flume schematic showing the setup of the flume and sampling locations for suspended sediment and gravel-stored sediment

was wet sieved which resulted in 2.7±0.1 kg of fine-grained sediment (oven-dry weight) being added to each of the three flume treatments.

The fine-grained sediment was delivered in four 15-L aliquots spaced 15 min apart to a stock container at the head of the flume (Fig. 1) that had a 800-cm$^2$ grid of 0.6-cm holes in the front and rear central portion of the container. The downstream grid was screened with 200 μm Nitex mesh (Sefar Inc., QC, Canada) to prevent particles >200 μm from entering the channel (similar to the method used by Rex and Petticrew 2010). The rapid but finite addition of sediment, as described above, may be characteristic of a channel bank failure or a small mass wasting event; channel banks have been identified as a common source of fluvial sediment (e.g., Walling 2005; Koiter et al. 2013b).

## 2.3 Sampling and analysis

The flume simulations were run for 31 h, corresponding to approximately 24 km of total distance traveled. The first samples were collected 15 min after the last sediment addition (i.e., 1 h after the initiation of the flume simulation) to ensure complete mixing within the flume environment. Suspended sediment samples (1 L) were collected using two Isco automatic water samplers (model 3700; Teledyne Isco, Inc, NE, USA) located at 9 and 27 m (Fig. 1) with the intake at 50 % of the flow depth. The suspended sediment samples were split and measured for particle size distribution, suspended sediment concentration, and organic matter content ("Particle size and organic matter content" section). Bulk (20 L) suspended sediment samples, for geochemical analysis, were collected mid-flume at 18 m at 1, 2, 4, 7, and 13 h, and these samples were allowed to settle at room temperature (~20 °C) for 48 h and the clear supernatant was decanted and the sediment samples were air-dried.

No bed sediment was collected for the 0 cm of gravel treatment as patterns of deposited sediment were assessed visually. A total of 12 sediment traps were used to collect gravel-stored sediment for the 5- and 40-cm gravel treatments. The sediment traps were placed in six rows, and within a row, traps were spaced equidistant from each other and the sidewalls of the flume. Each row was 3 m apart beginning 9 m from the front of the flume (Fig. 1). For the 5-cm gravel channel bed, gravel-stored sediments were collected using a sediment trap consisting of a plastic disk (35 cm diameter), with a small lip around the edge (1 cm high) which was placed on the bottom of the flume and covered with 5 cm of gravel. For the 40-cm gravel channel bed, gravel-stored sediments were collected using sediment traps that consisted of a waterproof bag (20 cm diameter and 35 cm long) secured to a metal ring and buried 20 cm deep into the gravel bed and covered with gravel (Rex and Petticrew 2008). At the end of the flume simulation run, the sediment traps were collected and the sample (including gravel) was placed in a 125-μm sieve and the fine-grained sediment separated from the gravel by rinsing with water. A subsample of the fine-grained sediment was taken for particle size analysis and organic matter content ("Particle size and organic matter content" section), with the remainder being allowed to settle. The clear supernatant was then decanted and the sediment was oven-dried and weighed.

### *2.3.1 Particle size and organic matter content*

The SSA of the sediment samples were measured using a Malvern Mastersizer 3000 laser diffraction system (Malvern, UK) (0.01–3500 μm diameter measurement range) assuming a constant particle density of 2.65 g cm$^{-3}$. The SSA of sediment samples was used as the primary measure of particle size as opposed to median particle size because comparisons of the median particle size would require that the distributions have similar shapes (e.g., normal distribution). However, it is important to note that increases in SSA correspond to a reduction in the median particle size diameter ($D_{50}$). The added, suspended, and gravel-stored sediment were measured for the non-dispersed or effective SSA (E-SSA) and the dispersed SSA (D-SSA), the latter following 2 min of in-line sonication at 90 % power (45 W). The suspended sediment samples were filtered using pre-ashed glass-fiber filters (0.7 μm pore size) to determine the suspended sediment concentration and ashed at 550 °C for 1 h to determine the organic matter content. Suspended sediment concentrations were corrected using a dilution factor to account for the additional pore water within the gravel bed matrix. The dilution factors were calculated using the measured gravel porosity and the depth of the gravel bed and were 1, 1.07, and 1.53 for the 0-, 5-, and 40-cm gravel bed treatments, respectively. Similarly, subsamples of the added sediment and gravel-stored sediment were ashed to determine organic matter content.

The bulk suspended sediment samples were analyzed for a broad suite of geochemical elements (51 elements in total) using ICP-MS following a microwave-assisted digestion with aqua regia (ALS Mineral Division, North Vancouver, BC, Canada). Subsamples of the bulk suspended sediment were also measured for the absolute SSA (A-SSA) following digestion with 30 % hydrogen peroxide to remove organics and disaggregated chemically with a solution of sodium hexametaphosphate and sodium carbonate (Kroetsch and Cang 2007) and physically with 2 min of sonication at 45 W (Misonix S-4000; Qsonica, Newtown, CT, USA).

## 2.4 Data analysis

All statistical analysis was undertaken using R Statistical Software v3.1.2 (R Core Team 2015) through RStudio Integrated Development Environment v0.98.5 (RStudio 2015). All plots were created using the R package ggplot2 v0.9.3.1 (Wickham 2009) with non-transformed data. For the suspended sediment, the change in concentration, organic matter content, and SSA over time, as well as differences between the three channel bed conditions, were assessed using linear regression with treatment contrasts using the 5-cm gravel treatment as the reference level. The relation between organic matter content and SSA for suspended sediment was non-linear and a piecewise linear regression was used. The data were split based on the largest SSA for the 0-cm gravel treatment which corresponded to 990.2 and 1112.4 $m^2\ kg^{-1}$ for the E-SSA and D-SSA, respectively.

Locally weighted regression, or LOESS, curves were used to help visualize the spatial patterns for the bed sediment characteristics and were generated using the stat_smooth function, and a spanning coefficient of 1.25 was used to prevent overfitting.

The relation between geochemical concentrations and A-SSA for the bulk suspended sediment samples was assessed using linear regression. A hypothetical size-corrected concentration regression line was created using the common correction factor which was based on the ratio of A-SSA of suspended to the added sediment (Collins et al. 1997). Using the regression line for the measured suspended sediment geochemical concentrations, it was assumed that at 965 $m^2\ kg^{-1}$, where the suspended and the original added sediment A-SSA are equal (i.e., correction factor=1.00), the geochemical concentrations were also equal. The concentration at this A-SSA was then multiplied by the calculated correction factors and plotted as the hypothetical size-corrected geochemical concentration.

## 3 Results

### 3.1 Changes in concentration, particle size, and organic matter content

#### *3.1.1 Suspended and gravel-stored sediment concentrations*

Given the total mass of the added sediment to the volume of water in the flume (excluding the volume of water contained within the gravel bed), the calculated maximum suspended sediment concentration, if all sediment remained in suspension with no mixing with gravel pore water, was approximately 170 mg $L^{-1}$ for each flume treatment. The suspended sediment concentrations were averaged between the two sampling locations (located at 9 and 27 m; Fig. 1) prior to statistical analysis, and the changes in the suspended sediment concentration over time are shown in Fig. 2a. The maximum recorded suspended sediment concentration (1 h) was 92.3, 90.5, and 57.5 mg $L^{-1}$ for the 0-, 5-, and 40-cm gravel bed treatments, respectively. Overall, the concentration decreased over time (Table 1a) and was characterized by a rapid decrease followed by a slow but continuous decline in concentration. At the termination of each flume simulation (31 h), the suspended sediment concentration was 25.7, 13.2, and 1.4 mg $L^{-1}$ corresponding to approximately 12.1, 6.0, and 0.6 % of the total added sediment remaining in suspension for the 0-, 5-, and 40-cm gravel bed treatments, respectively (Fig. 2a). There was a significant interaction between sampling time and channel bed conditions. The rate of decline in concentration was the lowest for the 0-cm gravel treatment followed by the 5-cm gravel treatment, and the rate of decline was the highest for the 40-cm gravel treatment (Table 1a).

No measurement of gravel-stored sediment was made for the 0-cm gravel treatment; however, visual observations showed that the amount of sediment on the bottom of the flume increased towards the back of the flume (i.e., the majority of sediment accumulated between 25 and 36 m; Fig. 1). The amounts of gravel-stored sediment for the 5- and 40-cm gravel beds are shown in Fig. 3a. There is a small difference between the two treatments with an average of 28.1±5.4 (±1 SD) and 35.6±5.4 g $m^{-2}$ for the 5- and 40-cm gravel treatments, respectively. There was no consistent trend in the amount of gravel-stored sediment along the length of the flume; however, there was evidence of a difference along the width (i.e., left to right) of the flume. This was especially evident in the 5-cm gravel treatment near the front (9–12 m; Fig. 3a) of the flume, and these observations suggest that the flow patterns were not linear along the entire length of the flume.

#### *3.1.2 Particle size*

The added sediment had an average E-SSA and D-SSA of 312.3±89.7 (± range) ($D_{50}$ 19.9±8.1 μm) and 450.3±78.1 $m^2\ kg^{-1}$ ($D_{50}$ 11.9±2.4 μm), respectively. The D-SSA was larger than the E-SSA due to the effect of particle aggregation. The SSA data for the suspended sediment were averaged between the two sampling locations (9 and 27 m; Fig. 1) prior to statistical analysis, and the E-SSA data are shown in Fig. 2b. The changes in the particle size had an inverse trend, but a similar rate of change, to the suspended sediment concentration. Overall, there was a significant increase in the SSA (i.e., decrease in particle size) over time (Table 1b), and it was characterized by an immediate and large increase in E-SSA following the addition of sediment as observed during the first suspended sediment measurement (1 h) with an E-SSA of 515.3, 586.2, and 630.2 $m^2\ kg^{-1}$ ($D_{50}$ 8.62, 7.13, and 6.24 μm) for the 0-, 5-, and 40-cm gravel treatments, respectively. There was an initial rapid increase in the E-SSA over the first few hours and then the rate of change slowed as the flume simulation progressed. For the 40-cm gravel treatment, there was insufficient suspended sediment after 11 h to make reliable particle size measurements. There was no significant interaction between particle type (E-SSA or D-SSA) and

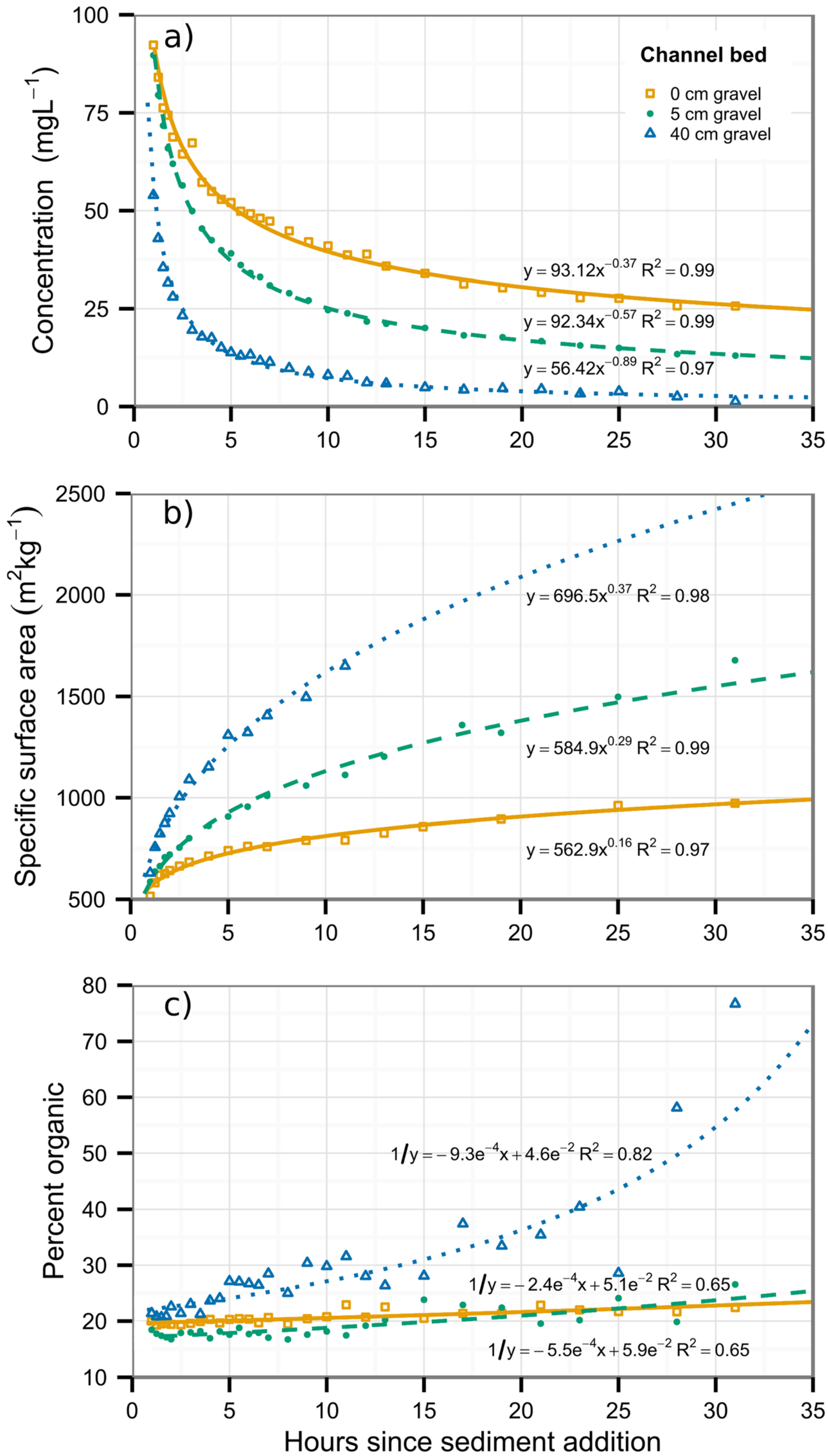


**Fig. 2** The change in suspended sediment concentration (corrected for dilution) (**a**), specific surface area (calculated using E-SSA) (**b**), and organic matter content of suspended sediment (**c**), with time (distance traveled) over three channel bed conditions. Dilution factors of 1, 1.07, and 1.53 were determined for the 0-, 5-, and 40-cm gravel beds, respectively

sampling time (h) (i.e., the relations between E-SSA or D-SSA and sampling time had similar characteristics), and this interaction term was removed from the analysis. The rate of increase in SSA was less for the 0-cm compared to the 5-cm gravel bed and the rate of increase in SSA was greater for 40-cm compared to the 5-cm gravel bed (Table 1b).

The E-SSA and D-SSA for the gravel-stored sediment for both 5- and 40-cm gravel beds was coarser than the added sediment (shown in Fig. 3b). The gravel-stored sediment is coarser in the 40-cm gravel treatment as compared to the 5-cm gravel treatment. Both the E-SSA and D-SSA showed a similar amount of variation with a range in the E-SSA of 69.2 and

**Table 1** Results of linear models assessing the correlation in concentration (a), specific surface area (b), and organic matter content (c) of the suspended sediment with hour (distance traveled), channel bed treatment, and particle type

| Analysis | Parameter | Estimate | Std. error | *df* | *t* value | $P(>\|t\|)$ |
|---|---|---|---|---|---|---|
| (a) Concentration $\log_{10}$ (mg $L^{-1}$) | (Intercept) | 1.965 | 0.018 | 81 | 109.34 | <0.001** |
| | $\log_{10}$ (h) | −0.566 | 0.019 | 81 | −29.41 | <0.001** |
| | 0 vs. 5 cm gravel | 0.004 | 0.025 | 81 | 0.14 | 0.887 |
| | 40 vs. 5 cm gravel | −0.214 | 0.025 | 81 | −8.42 | <0.001** |
| | $\log_{10}$ (h)×channel bed (1) | 0.193 | 0.027 | 81 | 7.11 | <0.001** |
| | $\log_{10}$ (h)×channel bed (2) | −0.327 | 0.027 | 81 | −12.01 | <0.001** |
| (b) Specific surface area $\log_{10}$ ($m^2$ $kg^{-1}$) | (Intercept) | 2.827 | 0.004 | 91 | 646.21 | <0.001** |
| | $\log_{10}$ (h) | 0.278 | 0.005 | 91 | 57.33 | <0.001** |
| | 0 vs. 5 cm gravel | −0.019 | 0.006 | 91 | −3.20 | 0.002* |
| | 40 vs. 5 cm gravel | 0.072 | 0.006 | 91 | 11.36 | <0.001** |
| | Particle type (E-SSA vs. D-SSA) | −0.055 | 0.003 | 91 | −20.37 | <0.001** |
| | $\log_{10}$ (h)×channel bed (1) | −0.123 | 0.007 | 91 | −17.92 | <0.001** |
| | $\log_{10}$ (h)×channel bed (2) | 0.088 | 0.009 | 91 | 9.43 | <0.001** |
| (c) Organic matter content $1/(\%_{organic})$ | (Intercept) | 0.0588 | 0.0009 | 81 | 63.38 | <0.001** |
| | Hour | −0.0006 | 0.0001 | 81 | −7.97 | <0.001** |
| | 0 vs. 5 cm gravel | −0.0078 | 0.0013 | 81 | −5.92 | <0.001** |
| | 40 vs. 5 cm gravel | −0.0126 | 0.0013 | 81 | −9.61 | <0.001** |
| | h×channel bed (1) | 0.0003 | 0.0001 | 81 | 3.23 | 0.002* |
| | h×channel bed (2) | −0.0004 | 0.0001 | 81 | −3.81 | <0.001** |

Channel bed contrasts compares 0 to 5 cm gravel (1) and 40 to 5 cm gravel (2)

*df* degrees of freedom

*$P<0.01$; **$P<0.001$

64.2 $m^2$ $kg^{-1}$ and a range in the D-SSA of 65.5 and 54.9 $m^2$ $kg^{-1}$ for the 5- and 40-cm gravel treatments, respectively. The E-SSA of the stored fine-grained bed sediment showed little change along the length of the flume (Fig. 3b).

### *3.1.3 Organic matter content*

The organic matter content of the added sediment ranged between 19.5 and 20.4 % with an average of 19.9 %. The organic matter contents of the suspended sediment were averaged between the two sampling locations (located at 9 and 27 m; Fig. 1) prior to statistical analysis, and the changes in organic matter content over time are shown in Fig. 2c. Initially, the organic matter of the suspended sediment was similar to the added sediment for all three treatments. Overall, as the flume simulations progressed, there was an increase in the suspended sediment organic matter content (Table 1c). At the termination of the flume simulation (31 h), there was only a small (<10 %) increase in the organic matter content compared to the start of the flume simulation (1 h) for the 0- and 5-cm gravel beds; however, there was a large increase in the organic matter content for the 40-cm gravel treatment. There was a significant interaction between sampling time and channel bed conditions. The rate of increase in organic matter content was less for the 0-cm gravel compared to the 5-cm gravel, and the rate of increase in organic matter content was greater for 40-cm gravel compared to the 5-cm gravel (Table 1c). The organic matter content of the bed sediment for the 5- and 40-cm gravel beds were both similar to the added sediment and show no spatial trends along the length of the flume with average values of 19.2±1.0 and 23.5±1.5 % for the 5- and 40-cm gravel treatments, respectively (Fig. 3c).

## 3.2 Relation between particle size and organic matter

The relation between organic matter content and E-SSA was non-linear (Fig. 4) over the range investigated. Overall, there was no significant relation for particles below the E-SSA threshold of 1000 $m^2$ $kg^{-1}$ and a significant positive relation for particles above the E-SSA threshold (Table 2). For the particles below the 1000 $m^2$ $kg^{-1}$ threshold, there was a significant interaction between E-SSA and channel bed conditions with the 0-cm gravel bed having a steeper positive slope compared to the 5-cm gravel bed (Table 2a). The relation between organic matter and E-SSA was the same for the 5- and 40-cm channel beds above the threshold value (Table 2b). There was no significant interaction between particle type (effective or dispersed sediment) and the SSA, and this interaction term was removed from the analysis.

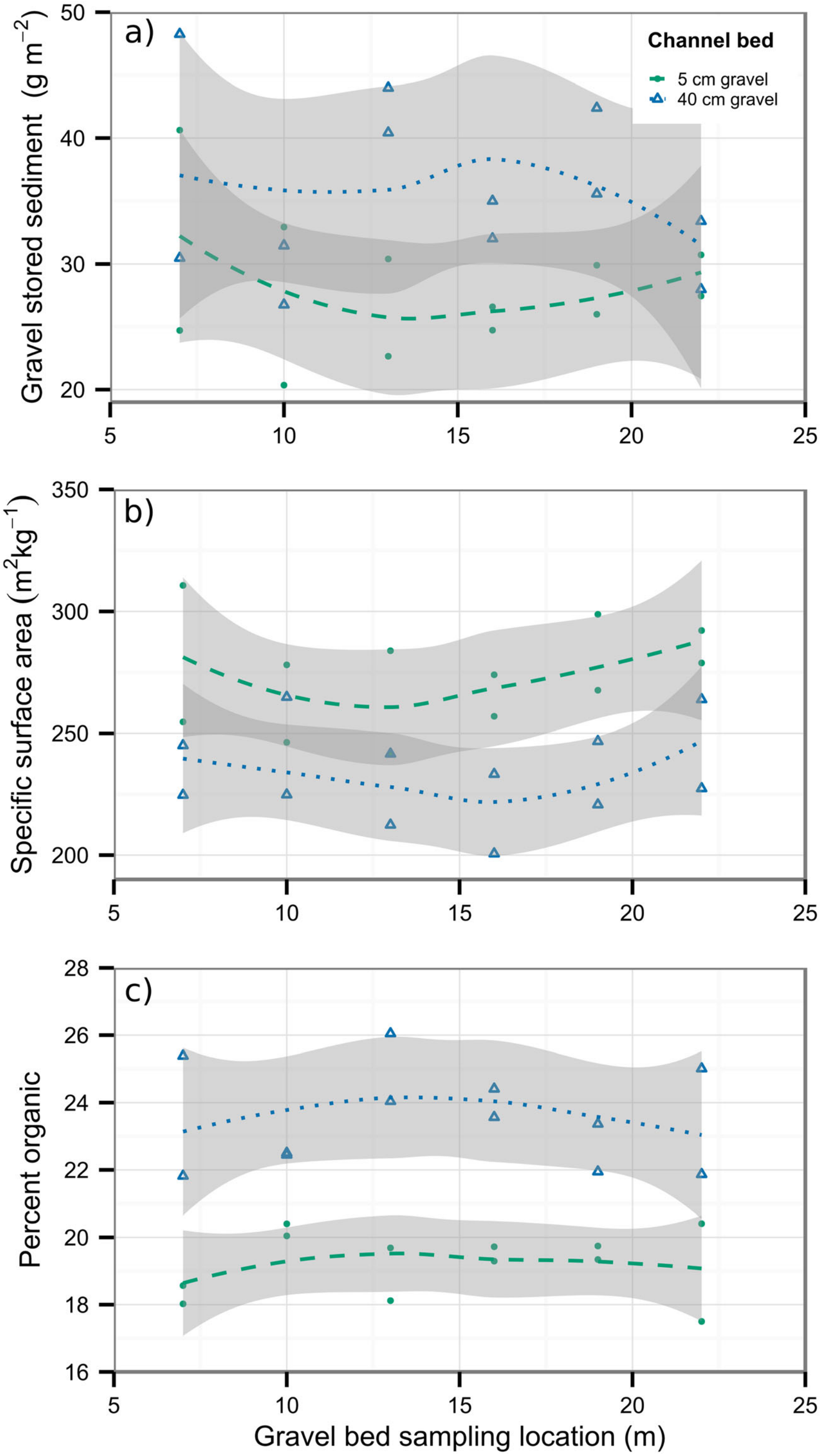


**Fig. 3** Properties of the gravel-stored fine-grained sediment along the length of the flume for the 5- and 40-cm gravel bed treatments. Sediment properties measured include **a** total stored mass, **b** specific surface area (calculated using E-SSA), and **c** organic matter composition. See Fig. 1 for sampling locations

### 3.3 Sediment geochemistry

Only the bulk suspended sediment samples collected in the 0-cm gravel bed treatment for the first 13 h had enough mass for geochemical analysis (0.5 g needed for analysis). The A-SSA of the added sediment was 959.3 m$^2$ kg$^{-1}$ ($D_{50}$ 4.21 μm) and the range of A-SSA for the suspended sediment was 964.7–1237.0 m$^2$ kg$^{-1}$ ($D_{50}$ 2.7–3.96 μm). The ratio between the A-SSA of the collected suspended sediment to the added sediment corresponds to a particle size correction factor ranging between 1.01 and 1.29 over the first 13 h of the flume simulation. The relation between geochemical concentration and SSA was not consistent between the different elements in terms of linearity and the direction and magnitude of the slope.

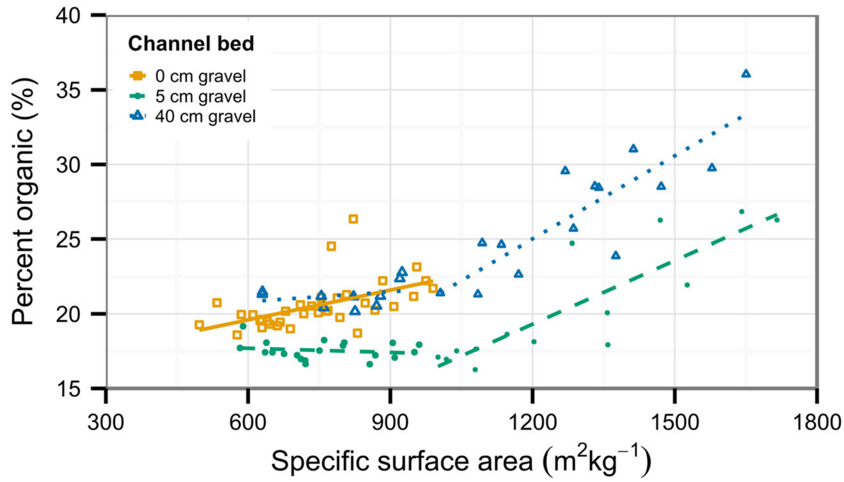


**Fig. 4** The relation between specific surface area (calculated using E-SSA) and organic matter content for the suspended sediment over three channel bed conditions

Out of the 51 elements investigated, six elements were below detection limits in one or more of the sediment samples and were removed from the analysis. Of the remaining 45 elements, 16 elements had a positive slope and two elements had negative slopes that were significantly different than 0 ($P<0.1$; $n=5$). Figure 5 shows examples of a positive (As), negative (La), and no clearly defined relation (Cd) between A-SSA and geochemical concentration. The hypothetical size corrected concentration is also plotted to highlight some of the potential errors associated with size-corrected geochemical concentrations. For example, at 13 h where the A-SSA of the suspended sediment was 1237 $m^2\ kg^{-1}$, there is the potential of an overestimation of the concentration of As and La of 9.5 and 43.0 %, respectively.

# 4 Discussion

## 4.1 Suspended sediment concentration and particle size selectivity

The effective particle size is typically a more relevant measure of particle size when investigating fluvial transport processes as it is the overall size, along with shape, composition, density, and internal structure of sediment particles, that determines the settling and entrainment velocities (Droppo 2001). However, results showed that the patterns of change in the particle size distribution over time (i.e., distance traveled) did not significantly differ between the E-SSA and D-SSA (Table 1b). This suggests that the particles were transported

**Table 2** Results of piecewise regression assessing the correlation in organic matter content for small (a) and large SSA (b) with SSA, channel bed treatment, and particle type for suspended sediment

| Analysis | Parameter | Estimate | Std. error | *df* | *t* value | $P(>\|t\|)$ |
|---|---|---|---|---|---|---|
| (a) Organic matter content (%) | (Intercept) | 17.832 | 1.141 | 125 | 15.63 | <0.001 |
| Small specific surface area | SSA | 0.000 | 0.001 | 125 | −0.09 | 0.932 |
| (<1000 $m^2\ kg^{-1}$) | 0 vs. 5 cm gravel | −2.099 | 1.357 | 125 | −1.55 | 0.124 |
| | 40 vs. 5 cm gravel | 1.534 | 2.119 | 125 | 0.72 | 0.471 |
| | Particle type (E-SSA vs. D-SSA) | −0.377 | 0.208 | 125 | −1.81 | 0.073 |
| | SSA×channel bed (1) | 0.006 | 0.002 | 125 | 3.88 | <0.001 |
| | SSA×channel bed (2) | 0.003 | 0.002 | 125 | 1.03 | 0.306 |
| (b) Organic matter content (%) | (Intercept) | 2.841 | 2.203 | 51 | 1.29 | 0.203 |
| Large specific surface area | SSA | 0.014 | 0.002 | 51 | 8.53 | <0.001 |
| (>1000 $m^2\ kg^{-1}$) | 40 vs. 5 cm gravel | 2.444 | 3.362 | 51 | 0.73 | 0.471 |
| | Particle type (E-SSA vs. D-SSA) | −2.514 | 0.603 | 51 | −4.17 | <0.001 |
| | SSA×channel bed (2) | 0.002 | 0.002 | 51 | 1.01 | 0.317 |

Channel bed contrasts compares 0 to 5 cm gravel (1) and 40 to 5 cm gravel (2)

*df* degrees of freedom

*$P<0.001$

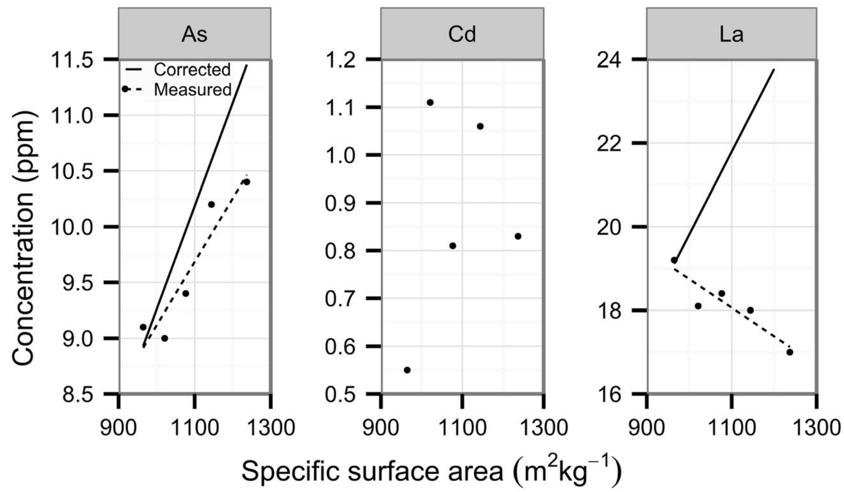


**Fig. 5** Three examples of the different relations between geochemical concentration and specific surface area (calculated using A-SSA), including the hypothetical size corrected concentration. Arsenic (*As*), positive relation ($p<0.1$); Cadmium (*Cd*), nonlinear relation; Lanthanum (*La*), negative relation ($p<0.1$)

as dense water-stable aggregates with similar hydrodynamic behavior as compared to discrete particles. However, it is unclear whether the stability of the aggregates reflects the properties of the added sediment, the flow characteristics within the flume, or the duration of the experiment. To assess whether flocculation was occurring along the length of the flume, the difference for both E-SSA and D-SSA between the two sampling locations (9 and 27 m; Fig. 1) was calculated for each treatment. There was not a significant ($P<0.05$) difference in either the E-SSA or D-SSA between the 9 and 27 m suspended sediment sampling locations (data not shown). Overall, there was a very small average difference in SSA between the two sampling locations of 15.1 and 1.9 $m^2$ $kg^{-1}$ for the E-SSA and D-SSA, respectively. The increase in both the E-SSA and D-SSA suggests there is some small degree of disaggregation or selective deposition of larger particles between the two sampling locations. The idea of selective deposition is supported by the small decrease in suspended sediment concentration (average difference of 0.4 mg $L^{-1}$) between the two sampling locations.

Regardless of channel bed conditions, there was a significant reduction in suspended sediment concentration and a decrease in the particle size and an increase in the organic matter content of the suspended sediment over time (i.e., distanced traveled). The presence of lower water velocities along the walls of the flume, due to the frictional resistance, and the greater depth of water and corresponding lower velocities at the rear of the flume will have played a role in promoting sediment deposition of the coarser sediment particles. Furthermore, the cohesive nature of the added sediment would have reduced the re-entrainment of deposited sediment. The Shields (1936) diagram shows that for inorganic particles < 200 μm, higher velocities and shear stress thresholds are needed for entrainment as the particle size decreases. For example, the median diameter (effective) of the added sediment was 20 μm and the corresponding lower limit of entrainment, according to the Hjulström (1935) curve, is approximately 40 cm $s^{-1}$ compared to the velocities of 20–25 cm $s^{-1}$ in this study.

Comparing the 0- to the 5-cm gravel bed treatments presents an opportunity to look at the effect of channel bed roughness and gravel bed pore volume on the preferential trapping of sediment. The addition of 5 cm of gravel significantly reduced the overall suspended sediment concentration, resulted in smaller particle sizes, and increased the organic matter content of the suspended sediment compared to the 0-cm gravel bed. The roughness elements created by the gravel changes the vertical velocity profile by slowing the water velocity near the bed surface due to drag forces (increased fluid contact at the channel bed boundary) (Kirkgöz 1989). Manning's roughness coefficient for a straight uniform concrete channel ranges between 0.012 and 0.018 as compared to 0.028–0.035 for a gravel bed (Arcement and Schneider 1989). The resulting reduction in near bed velocity would allow for smaller grains of sediment to settle. In many natural settings, the channel bed is not composed of grains of a single size class (uniform) but rather composed of a wide range in particle size from clay and silts to coarse sands and gravels. The entrainment of particles from non-uniform beds tend not to follow the Shields diagram, and fine-grained sediments of non-uniform channel beds are more difficult to entrain compared to the same sized particles in a uniform channel bed (Xu et al. 2008). Within the flume simulation, the addition of a gravel bed created a bed with a bimodal grain size distribution (i.e., the added fine-grained sediment and the coarse gravel), and the gravel would have acted as a protecting layer preventing the erosion of the fine-grained sediment. Furthermore, the addition of the gravel would have increased the amount of turbulence near the channel bed. Turbulence can penetrate below the channel surface and can result in the resuspension of sediment stored near the channel bed surface (Packman and Salehin 2003). However, this effect diminishes with depth and may, in part, explain the lower concentrations and smaller particles in the 40-cm as compared to the 5-cm gravel beds.

The comparison of the 40- to the 5-cm gravel channel bed allows for the investigation of the effects of intra-gravel flow on the dynamics of suspended sediments. Previous research (Slager 2014), using salt as a tracer in a similar QRRC flume, under similar flow and gravel bed conditions, showed that the water residence time quickly increased with depth. The residence time within the gravel matrix was approximately 5 and 55 times greater at a depth of 5 and 15 cm, respectively, compared to the channel bed surface (Slager 2014). Intra-gravel flows exhibit a sieve-like removal of all but the very fine-grained sediment as the tortuous flow pathways though the gravel would slow the velocity of the flow allowing for a greater amount of sediment deposition. For example, Petticrew et al. (2007) demonstrated through the use of lidded and non-lidded channel bed sediment traps that a significant amount of fine-grained sediments can be deposited by lateral flows through gravel. With larger grain sizes (e.g., sand), particles can become trapped among the pore spaces near the surface of the gravel creating a seal, or cap, preventing the downward migration of sediment; however, given the fine-grained nature of the added sediment and the high porosity of the gravel in this study, it is likely that the added sediment would penetrate deep into the gravel bed. The fine-grained sediment stored deeper in the gravel bed is less exposed to turbulence, which would limit the amount of sediment being resuspended (Gibson et al. 2011; Hamm et al. 2011).

The nature of the sediment supply is important to consider as it will influence the particle size and organic matter selectivity of fluvial transported sediment. For example, a sediment pulse, as demonstrated in this study, will be different as compared to a more continuous supply of sediment (e.g., surface erosion). The latter would maintain a higher and more consistent suspended sediment concentration resulting in a higher fluid density which will change the settling velocity of sediment particles. In addition, the continuous deposition of sediment will eventually exceed the storage capacity of the gravel bed or create a surface seal changing the boundary conditions from a rough and porous to a smooth and impermeable surface, similar to the 0-cm gravel treatment.

### 4.2 Organic matter selectivity

All three flume treatments show an increase in the organic matter content of the suspended sediment over time. This is likely due, in part, to the difference between the density of organic matter (0.9–1.3 Mg $m^{-3}$) and mineral particles (2.6–2.75 Mg $m^{-3}$) (Brady and Weil 2001). The decrease in density generally lowers the settling velocity allowing for the preferential transport of organic-rich particles. The aggregated sediment particles may have been breaking apart due to the pumps within the recirculating flume setup and breaking down the aggregates into their primary organic and inorganic components. The more dense mineral fraction preferentially settles while the organic component remains in suspension which results in the organic-rich sediment remaining in suspension.

The relation between E-SSA and organic matter content of the suspended sediment was not linear over the range of E-SSA investigated. Only suspended sediment particles over an E-SSA threshold value of 1000 $m^2$ $kg^{-1}$ showed a significant positive relation between E-SSA and organic matter content. This suggests that the selective process involved in fluvial transport is different, in terms of magnitude of the effect and the trend over time, between particle size and organic matter. This was also evident with respect to changes over time for each property as particle size had a log-log relation (Fig. 2b) while organic matter had a reciprocal relation (Fig. 2c). Furthermore, making generalizations about the organic matter content based on particle size may depend on the range of E-SSA being investigated, i.e., the relation may be considered linear over small ranges of E-SSA. Small sample masses prevented the investigation of the relation between A-SSA and organic matter which is needed to fully understand the particle size and organic matter dynamics.

### 4.3 Implications for the geochemical composition of suspended sediment

The lack of any consistent trend between the different geochemical elements may suggest that the mineralogy of the inorganic fraction of the sediment is not uniform across particle sizes (Fig. 5). The distribution of geochemical elements within sediment, whether it is part of the crystalline structure (e.g., primary or secondary minerals) or absorbed onto the surface (e.g., exchangeable), may affect the relation between A-SSA and geochemical concentrations. Other sediment properties may also influence the A-SSA and geochemical concentration relation, for example, Fe-oxides have been shown to increase soil aggregation (e.g., Duiker et al. 2003) as well as have a large influence on the geochemical composition of soils due to their high A-SSA (Horowitz 1991). Therefore, a larger single aggregate will have a lower E-SSA than the sum of the A-SSA for all the individual particles that the aggregate is composed of. When these larger aggregates are preferentially deposited, they may sequester a disproportional amount of other elements compared to the smaller aggregates or discreet particles remaining in suspension. The dissolution and desorption of elements from the added sediment once entering the flume environment would have also contributed to the range of trends observed. The organic matter content of the sediment may have also influenced the geochemical composition; however, the added effects of organic matter may have been limited as there was only a small change (1–13 h) in organic matter content for the 0-cm gravel bed

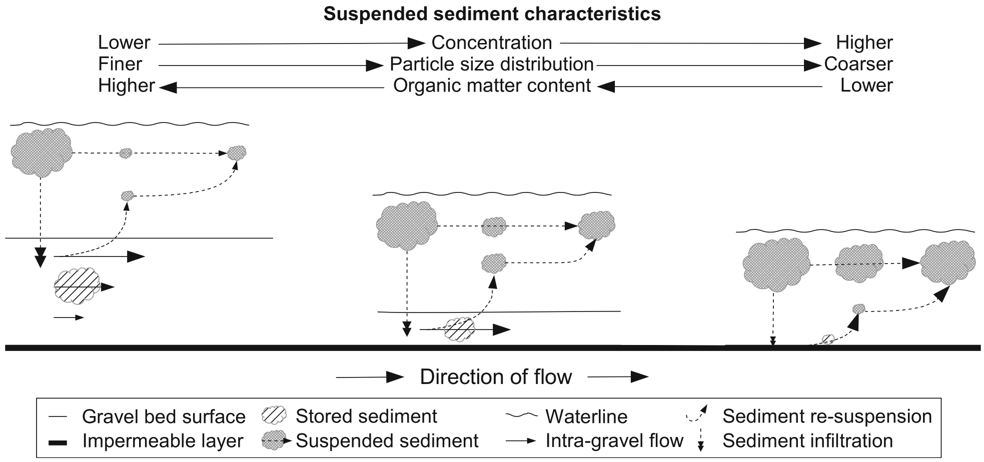


**Fig. 6** Conceptual diagram showing the influence of gravel beds on the deposition, storage, and resuspension of fine-grained sediment. The gravel bed increases bed roughness slowing the velocity near the channel bed which promotes deposition. Increasing the depth of the gravel bed modifies the intra-gravel flow resulting in greater storage and limiting the resuspension of sediment

treatment for which the geochemistry of the suspended sediment was investigated (Fig. 2b). Further investigation of the partitioning of geochemical elements into various fractions (e.g., exchangeable, bound to organic matter, and residual) is needed to fully understand the relation between A-SSA, organic matter content, and geochemical concentrations.

This study highlights the importance of the particle size selectivity process on geochemical (e.g., trace metals) flux and transport studies. It is important to establish the relations between particle size and elemental concentrations for each element of interest as the results from this study show that the relation between particle size and elemental concentration can vary between elements. Once the particle size and elemental concentration relation has been established, this information can be used in conjunction with both the suspended sediment concentration and particle size distribution to estimate total load. Furthermore, the results from this study demonstrate that both the particle size and organic matter content selectivity can change according to the channel bed conditions, specifically the channel bed roughness and the presence of high porosity gravels. Figure 6 depicts a conceptual diagram showing the changes in particle size, organic matter content, suspended sediment concentration, and gravel bed sediment storage with changing gravel bed depth as observed in this study. This is important to consider as the channel bed conditions can change over time from high flow events that flush out gravel-stored fine-grained sediment or result in gravel bed movement. In addition, channel bed condition can also vary longitudinally from headwaters towards the watershed outlet as channels transition between bed types, for example, from bedrock to gravel through to sand channel bed types. The spatial and temporal variations in particle size and organic matter selectivity need to be considered when selecting sampling locations and timing and interpreting the data (Koiter et al. 2013a).

The relation between particle selectivity and geochemical composition is an important aspect of many sediment fingerprinting studies. Typically, there is a difference in both the particle size distribution and organic matter content between potential sources of sediment and downstream collected sediment that is adjusted for by using simple correction factors based on ratios of A-SSA and organic matter content between source and sediment. However, Smith and Blake (2014) demonstrated that the particle size and organic matter correction factors can have a large influence in the final source apportionment results. Therefore, it is important to test the validity of such correction factors prior to using them. The results from this study, although limited, support the conclusions of Koiter et al. (2013a) for the need to carefully consider particle size correction factors and the elements selected as tracers in sediment fingerprinting studies, as not all elements have the same A-SSA and concentration relation. The range in relations found between the different geochemical elements in this study is similar to the research by others (e.g., Russell et al. 2001; Smith and Blake 2014) and draws attention to the assumption made when using particle size correction factors. This research highlights the continued need for research and empirical evidence for the processes of particle size and organic matter selectivity and their subsequent influence on geochemical properties if the sources and sinks are going to be linked in a robust and reliable manner. Fluvial transport is one of the last steps of the sediment cascade, and further work is needed to investigate the additional influences of particle selectivity of the erosional and sediment delivery processes when linking sediment sources to downstream collection.

## 5 Conclusions

Using a recirculating flume, it was demonstrated that the distanced traveled (i.e., transport time) and channel bed characteristics both had a significant effect on the particle size and organic matter selectivity of the transported suspended sediment. Furthermore, it was shown that a gravel bed (0- vs. 5-cm gravel treatments) introduced small-scale roughness which resulted in the preferential deposition of larger particles into the channel bed as compared to a smooth planar channel bed. The change from the 5- to 40-cm gravel bed treatments increased the amount of intra-gravel flow and reduced the potential for resuspension, resulting in further reduction in the particle size of the suspended sediment. This information demonstrates that the channel bed conditions are an important factor to consider when selecting sediment sampling sites and interpreting the data they provide. The geochemical properties of the suspended sediment changed as a result of fluvial transport, but it was difficult to properly account for these changes based on particle size alone. There are other confounding factors, including the relation of aggregate size and stability to sediment geochemistry, that need to be considered. This research helps to understand the process of particle selectivity, in terms of particle size and organic matter content, which is an important part of many sediment management tools, as well as to aid in the interpretation of sediment-associated nutrient and contaminant transport dynamics.

**Acknowledgments** The first author is supported by the Natural Science and Engineering Research Council (NSERC) of Canada through the Alexander Graham Bell Canada Graduate Scholarship Program. This work is funded by a NSERC Strategic Grant—Development of environmental fingerprinting techniques for sources of sediment and associated phosphorus within agricultural watersheds of Canada—to DAL, PNO, and ELP. The authors would also like to acknowledge the Quesnel River Research Centre (QRRC) and the Landscape Ecology Research Group staff and students for the use of the facilities and equipment as well as technical support. This paper comprises part of the QRRC Publication Series.

## References

Albers SJ, Petticrew EL (2012) Ecosystem response to a salmon disturbance regime: implications for downstream nutrient fluxes in aquatic systems. Limnol Oceanogr 57:113–123

Angers DA, Bullock MS, Mehuys GR (2007) Aggregate stability to water. In: Carter MR, Gregorich EG (eds) Methods of soil analysis, 2nd edn. CRC Press, Boca Raton, pp 811–819

Arcement GJ, Schneider VR (1989) Guide for selecting manning's roughness coefficients for natural channels and flood plains. Report 2339. U.S. Department of Transportation, Federal Highway Administration, Reston

Bainbridge ZT, Lewis SE, Smithers SG, Kuhnert PM, Henderson BL, Brodie JE (2014) Fine-suspended sediment and water budgets for a large, seasonally dry tropical catchment: Burdekin River catchment, Queensland, Australia. Water Resour Res 50:9067–9087

Brady NC, Weil RR (2001) The nature and properties of soils, 13th edn. Prentice Hall, New Jersey

Cole JJ, Prairie YT, Caraco NF, McDowell WH, Tranvik LH, Striegle RG, Duarte CM, Kortelainen P, Downing JA, Middleburg JJ, Melack J (2007) Plumbing the global carbon cycle: integrating inland waters into the terrestrial carbon budget. Ecosystems 10:172–185

Collins AL, Walling DE, Leeks GJL (1997) Source type ascription for fluvial suspended sediment based on a quantitative composite fingerprinting technique. Catena 29:1–27

Davis CM, Fox JF (2009) Sediment fingerprinting: review of the method and future improvements for allocating nonpoint source pollution. Environ Eng 135:490–504

Di Stefano C, Ferro V (2002) Soil and water: linking clay enrichment and sediment delivery processes. Biosyst Eng 81:465–479

Droppo IG (2001) Rethinking what constitutes suspended sediment. Hydrol Process 15:1551–1564

Droppo IG, Leppard GG, Flannigan DT, Liss SN (1997) The freshwater floc: a functional relationship of water and organic and inorganic floc constituents affecting suspended sediment properties. Water Air Soil Pollut 99:43–53

Duiker SW, Rhoton FE, Torrent J, Smeck NE, Lal R (2003) Iron (hydr)oxide crystallinity effects on soil aggregation. Soil Sci Soc Am J 67:606–611

Fryirs K (2013) (Dis)Connectivity in catchment sediment cascades: a fresh look at the sediment delivery problem. Earth Surf Process Landforms 38:30–46

Garzon-Garcia A, Olley JM, Bunn SE (2015) Controls on carbon and nitrogen export in an eroding catchment of south-eastern Queensland, Australia. Hydrol Process 29:739–751

Gibson S, Heath R, Abraham D, Schoellhamer D (2011) Visualization and analysis of temporal trends of sand infiltration into a gravel bed. Water Resour Res 47, W12601

Grams PE, Wilcock PR (2014) Transport of fine sediment over a coarse, immobile riverbed. J Geophys Res:Earth 119:188–211

Hamm NT, Dade WB, Renshaw CE (2011) Fine particle deposition to porous beds. Water Resour Res 47, W11508

Hjulström F (1935) Studies of the morphological activity of rivers as illustrated by the River Fyris. Bull Geol Instit Uppsala 25:221–527

Horowitz AJ (1991) A primer on sediment-trace element chemistry, 2nd edn. Lewis Publishers, Chelsea

Kerr JG, Burford MA, Olley JM, Bunn SE, Udy J (2011) Examining the link between terrestrial and aquatic phosphorus speciation in a subtropical catchment: the role of selective erosion and transport of fine sediments during storm events. Water Res 45:3331–3340

Kirkgöz MS (1989) Turbulent velocity profiles for smooth and rough open channel flow. J Hydraul Eng 115:1543–1561

Koiter AJ, Owens PN, Petticrew EL, Lobb DA (2013a) The behavioural characteristics of sediment properties and their implications for sediment fingerprinting as an approach for identifying sediment sources in river basins. Earth-Sci Rev 125:24–42

Koiter AJ, Lobb DA, Owens PN, Petticrew EL, Li S, Tiessen KHD (2013b) Investigating the role of connectivity and scale in assessing the sources of sediment in an agricultural watershed in the Canadian prairies using sediment source fingerprinting. J Soils Sediments 13:1676–1691

Kroetsch D, Cang C (2007) Particle size distribution. In: Carter MR, Gregorich EG (eds) Soil sampling and methods of analysis, 2nd edn. CRC Press, Boca Raton, pp 713–727

Laceby JP, McMahon J, Evrard O, Olley J (2015) A comparison of geological and statistical approaches to element selection for sediment fingerprinting. J Soils Sediments. doi:10.1007/s11368-015-1111-9

Maskaoui K, Zhou JL (2010) Colloids as a sink for certain pharmaceuticals in the aquatic environment. Environ Sci Pollut Res 17:898–907

Naden PS (2010) The fine-sediment cascade. In: Burt TP, Allison RJ (eds) Sediment cascades: an integrated approach. Wiley, Chichester, pp 271–305

Ockenden MC, Deasy C, Quinton JN, Surridge B, Stoate C (2014) Keeping agricultural soil out of rivers: evidence of sediment and nutrient accumulation within field wetlands in the UK. J Environ Manage 135:54–62

Oliver DM, Clegg CD, Heathwaite AL, Haygarth PM (2007) Preferential attachment of *Escherichia coli* to different particle size fractions of an agricultural grassland soil. Water Air Soil Pollut 185:369–375
Packman AI, Salehin M (2003) Relative roles of stream flow and sedimentary conditions in controlling hyporheic exchange. Hydrobiologia 494:291–297
Petticrew EL, Krein A, Walling DE (2007) Evaluating fine sediment mobilization and storage in a gravel-bed river using controlled reservoir releases. Hydrol Process 21:198–210
Pulley S, Foster I, Antunes P (2015) The application of sediment fingerprinting to floodplain and lake sediment cores: assumptions and uncertainties evaluated through case studies in the Nene Basin, UK. J Soils Sediments. doi:10.1007/s11368-015-1136-0
R Core Team (2015) R: A language and environment for statistical computing. R Foundation for Statistical Computing. http://www.R-project.org
Rehg KJ, Packman AI, Ren JH (2005) Effects of suspended sediment characteristics and bed sediment transport on streambed clogging. Hydrol Process 19:413–427
Rex JF, Petticrew EL (2008) Delivery of marine-derived nutrients to streambeds by Pacific salmon. Nat Geosci 1:840–843
Rex JF, Petticrew EL (2010) Salmon-derived nitrogen delivery and storage within a gravel bed: sediment and water interactions. Ecol Eng 36:1167–1173
Rhoton FE, Emmerich WE, Goodrich DC, Miller SN, McChesney DS (2006) Soil geomorphological characteristics of a semiarid watershed. Soil Sci Soc Am J 70:1532
RStudio (2015) RStudio: integrated development environment for R. http://www.rstudio.org/
Russell MA, Walling DE, Hodgkinson RA (2001) Suspended sediment sources in two small lowland agricultural catchments in the UK. J Hydrol 252:1–24
Sherriff SC, Franks SW, Rowan JS, Fenton O, Ó'hUallacháin D (2015) Uncertainty-based assessment of tracer selection, tracer non-conservativeness and multiple solutions in sediment fingerprinting using synthetic and field data. J Soils Sediments. doi:10.1007/s11368-015-1123-5
Shields A (1936) Anwendung der Aehnlichkeitsmechanik und der urbulenzforschung auf die eschiebebewegung. Mitteilung der preussischen Versuchsanstalt fur Wass erbau und Schiffba, Berlin
Slager A (2014) Hyporheic exchange inside a flat gravel bed, flume experiments and modelling. M.Sc. thesis, Utrecht University. Netherlands
Slattery MC, Burt TP (1997) Particle size characteristics of suspended sediment in hillslope runoff and stream flow. Earth Surf Process Landforms 22:705–719
Smith HG, Blake WH (2014) Sediment fingerprinting in agricultural catchments: a critical re-examination of source discrimination and data corrections. Geomorphology 204:177–191
Stone PM, Walling DE (1997) Particle size selectivity considerations in suspended sediment budget investigations. Water Air Soil Pollut 99: 63–70
Tranvik LJ, Downing JA, Cotner JB, Loiselle SA, Striegl RG, Ballatore TJ, Dillon P, Finlay K, Fortino K, Knoll LB (2009) Lakes and reservoirs as regulators of carbon cycling and climate. Limnol Oceanogr 54:2298–2314
Walling DE (1983) The sediment delivery problem. J Hydrol 65:209–237
Walling DE (2005) Tracing suspended sediment sources in catchments and river systems. Sci Total Environ 344:159–184
Walling DE, Webb BW, Woodward JC (1992) Some sampling considerations in the design of effective strategies for monitoring sediment-associated transport. In: Bogen J, Walling DE, Day TJ (eds) Erosion and sediment transport monitoring programmes in river basins. IAHS Publ. no. 210. IAHS Press, Wallingford, pp 279–288
Wickham H (2009) ggplot2: elegant graphics for data analysis. Springer, New York
Wilkinson SN, Olley JM, Furuichi T, Burton J, Kinsey-Henderson AE (2015) Sediment source tracing with stratified sampling and weightings based on spatial gradients in soil erosion. J Soils Sediments. doi:10.1007/s11368-015-1134-2
Withers P, Jarvie H (2008) Delivery and cycling of phosphorus in rivers: a review. Sci of Total Environ 400:379–395
Wooster JK, Dusterhoff SR, Cui Y, Sklar LS, Dietrich WE, Malko M (2008) Sediment supply and relative size distribution effects on fine sediment infiltration into immobile gravels. Water Resour Res 44, W03424
Xu H, Lu J, Liu X (2008) Non-uniform sediment incipient velocity. Int J Sediment Res 23:69–75
Young R, Huryn A (1997) Longitudinal patterns of organic matter transport and turnover along a New Zealand grassland river. Freshwater Biol 38:93–107
Zheng Y, Luo X, Zhang W, Wu B, Han F, Lin Z, Wang X (2012) Enrichment behavior and transport mechanism of soil-bound PAHs during rainfall-runoff events. Environ Pollut 171:85–92
Zingg T (1935) Beitrag zur schotteranalyse. Schweizerische mineralogische und petrologische mitteilungen 15:39–140