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The role of soil surface properties on the particle size and carbon selectivity of interrill erosion in agricultural landscapes

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ABSTRACT

The selective nature of interrill erosion – the preferential mobilization and transport of soil particles – can lead to the enrichment of fine-grained soil particles (<63 μm) and organic carbon (OC) within the mobilized soil. This study investigated the effects of slope gradient, vegetative cover, soil moisture content, texture and organic matter content, as well as their interactions on soil loss, particle size distribution and organic carbon content of soil mobilized under simulated rainfall within two contrasting agricultural regions in Canada. Overall, it was found that the eroded material was enriched in both fine-grained and carbon-rich particles relative to the source soil. It was demonstrated that dispersing and sieving both the source soil and the mobilized soil to <63 μm (i.e., removal of sand and large particulate organic matter) reduced the relative enrichment of both fine-grained soil particles and OC, which would allow for a more direct comparison of physical and biogeochemical properties between the source and mobilized soil. Furthermore, it was demonstrated that while the soil loss and the degree of enrichment were negatively correlated, there were differences in which soil surface properties had a significant effect in determining soil loss and the selectivity of both fine-grained and organic-rich soil particles and how these surface properties interacted. This suggests that while soil loss may be a good predictor of the degree of enrichment, the factors that control these two processes are different, which is important in understanding the process of selectivity. It was also shown that the OC content of both the source and mobilized soil was positively correlated to the degree of soil aggregation in addition to the silt and clay content.

1. Introduction

Soils are an important natural resource that are required for the majority of the global food and fibre production for human consumption and use. However, the growing global population has decreased the per capita arable land area and the productivity of this land is being diminished by degradation (Foley et al., 2005; Lal, 2006; Pimentel, 2006). One of the main causes of land degradation is soil erosion by water, wind and tillage (Lal, 2006; Montanarella et al., 2016). Soil erosion reduces crop yields through the loss of organic- and nutrient-rich topsoil. Furthermore, the sediment produced by soil erosion, when delivered to surface waterways, can result in the degradation in water quality through increased turbidity and nutrient loading (Bilotta and Brazier, 2008). In an effort to address these issues there has been a tremendous amount of research over the past century investigating the process of interrill erosion and its controlling factors (Dotterweich, 2013).

The rate of interrill erosion is often modelled as a function of the combined effects of the inherent erodibility of the soil, slope and the erosivity of the rainfall intensity (Elliot et al., 1987). Adjustment factors are often used to account for different site conditions, including canopy cover, ground cover, and sealing and crusting (Alberts et al., 1995). In contrast, there has been less research on the effects of these factors on the physical and biogeochemical properties of the mobilized, transported and deposited material despite the fact that soil erosion has been identified as a selective process in terms of both particle size and organic matter content (Ballantine et al., 2008; Chartier et al., 2013). For example, selectivity occurs when mobilized soil (eroded material) has a finer grain-size distribution and a higher organic matter content as compared to the source material (soil) as the smaller and less dense particles are preferentially mobilized and transported. However, the influence of properties at the soil surface (e.g., ground cover, slope gradient and soil texture) on the preferential mobilization of soil particles has not been fully evaluated.

The preferential mobilization and transport of organic-rich soil particles are important processes as organic carbon (OC) has a large impact on soil physical (e.g., water retention), chemical (e.g., nutrient

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retention), biological (e.g., biodiversity) and ecological (e.g., carbon sequestration) qualities (Lal, 2014). Interrill erosion and more generally soil erosion, is an important, albeit a poorly understood, component of the global carbon (C) cycle (Van Oost et al., 2007). The erosion of OC results in the redistribution of the total OC pool within the landscape at the field-scale, both laterally and vertically, resulting in a decline in OC in eroding areas and a corresponding increase in OC in depositional areas (Van Oost et al., 2007). Fine-grained and organic-rich soil particles can act as a vector for nutrients and other contaminants (Quinton and Catt, 2007; Oliver et al., 2007; Zheng et al., 2012; Yang et al., 2013). The higher concentrations of nutrients within the fine-grained and organic-rich fraction coupled with their preferential mobilization and transport results in a lateral and vertical redistribution of nutrients across the landscape increasing the environmental significance of erosion (Zhang et al., 2014). Soil erosion can also have an influence on other soil physical and chemical properties including pH, stone content, carbonate concentration and bulk density (Li et al., 2007). The net effect of soil erosion processes on the C balance and nutrient distribution depends on the scale of investigation which can range from plot- to field- to watershed-scale. Synthesizing information about the processes that regulate C and nutrient dynamics at different spatial scales will help clarify the role soil erosion has on the global C cycle. Information on how properties of the soil surface determine the organic matter selectivity of soil erosion will also provide additional predictive capabilities regarding C and nutrient dynamics in agricultural systems.

Particle size and organic matter selectivity and the resultant enrichment within the eroded material is primarily driven by the energy regime of the raindrop impacts and overland flow, with higher energy erosional processes being less selective (Proffitt and Rose, 1991; Issa et al., 2006; Schiettecatte et al., 2008; Armstrong et al., 2011). However, soil surface properties can also influence the process of selectivity. The influence of a range of soil surface properties including texture, organic matter content, vegetative cover and slope gradient on both particle size distribution and organic matter composition of eroded material have been investigated (Quinton et al., 2001; Armstrong et al., 2011; Defersha and Melesse, 2012; Shi et al., 2013; Chartier et al., 2013). Information on the selectivity of interrill erosion processes is needed as input for watershed management tools including sediment budgets and the assessment of geomorphic connectivity, as fine-grained and organic-rich particles can have different patterns of erosion, transport and deposition (Stone and Walling, 1997; Di Stefano and Ferro, 2002). Another important watershed management tool that is influenced by particle selectivity is sediment fingerprinting (Koiter et al., 2013). The sediment fingerprinting technique uses the physical or biogeochemical properties of soils as tracers, and is employed to identify sediment sources and estimate their contributions to the total sediment load in streams (Owens et al., 2016). However, many of the properties commonly used as tracers (e.g., trace metal concentrations and fallout radionuclide activities) are sensitive to changes in particle size distribution and organic matter content. Knowledge regarding the process of selectivity is needed to account for these changes so that a more direct comparison between source soils and sediment collected downstream can be made (Koiter et al., 2013; Smith and Blake, 2014).

The purpose of this research was to identify the significance of a range of naturally occurring soil surface properties in agricultural landscapes and their interactions on interrill erosion in terms of the enrichment of fine-grained particles and soil organic carbon (SOC). The soil surface properties examined include: vegetative cover, slope gradient, silt content, soil organic matter content and antecedent soil moisture content. The objectives of this research were: (1) to assess the enrichment of fine-grained particles (<63 μm) and SOC of mobilized soil with respect to their source soils; (2) to identify how properties of the soil surface influence soil loss and the enrichment of fine-grained particles and SOC in the mobilized soil; (3) to investigate the relation between SOC and particle size distribution (primary and aggregated) in both the source soil and the mobilized soil; and (4) to characterize the implications of the selectivity of interrill erosion for C, nutrient and geochemical fluxes.

2. Materials and methods

2.1. Site descriptions

Two contrasting agricultural regions in Canada were used to investigate particle size and organic matter selectivity of soil erosion under a range of soil surface properties (e.g., vegetative cover and texture). The first region was the South Tobacco Creek (STC) watershed, which is situated in south-central Manitoba (Fig. 1). The STC watershed is 75 km^2 and is predominately agriculture with the majority of land under annual crops including wheat, canola, flax and barley. The STC watershed extends across the Manitoba Escarpment; its upper reaches lie in undulating glacial tills and its lower reaches lie in the lacustrine sediments of glacial Lake Agassiz (Agriculture and Agri-Food Canada, 2011). The soils above the escarpment are Dark Grey Chernozems with a clay-loam texture, below the escarpment the soils are Black Chernozems with a clay texture and the valley walls soils are Regosols (Hope et al., 2002). The second region was the Beaver Valley (BV) watershed which is situated in the Fraser Plateau, located in central British Columbia (Fig. 1). The BV watershed is predominately forested with pasture and forage land uses in the flatter valley bottom sections. This study targeted a 10 km stretch of agricultural land in the valley bottom where the soils are Dystric Brunisols and Orthic Regosols, with a sandy loam texture with 5–10% coarse fragments (>2 mm) (Lord, 1984).

In each watershed, a series of hillslopes within areas of active agricultural production were identified as being degraded and/or having a high potential for soil erosion and a moderate to high degree of connectivity to surface waterways. The assessment of soil degradation was based primarily on vegetative cover (i.e., high grazing pressures or intensive crop production) and compaction and/or soil disturbance (i.e., farm vehicle and cattle tracks, and/or cultivation). This approach to site selection was taken as studies have shown that these areas can contribute a disproportionate amount of soil loss given their areal extent and are, therefore, of considerable interest within the context of the sediment cascade (Pietola et al., 2005; Raper, 2005; Collins et al., 2010). Within the STC watershed five hillslopes were identified that spanned the different physiographic regions of the watershed: above, within and below the Manitoba Escarpment. The hillslopes identified above and below the escarpment were cultivated fields and the hillslope within the escarpment was a forested site containing a cattle trail going down a steep valley wall towards the creek. Similarly, in the BV watershed a total of six hillslopes were identified with the disturbance ranging from winter feeding sites, areas with high grazing pressure and cattle and vehicle paths.

On each hillslope, a transect was established extending from the hilltop down to either the field or riparian edge. On each transect, rainfall simulation runoff plots were established on the upper, mid and lower slope positions. Different hillslope positions were used as this exploited the natural and contemporary patterns of soil erosion and served to expand the quantitative range of soil surface properties investigated. Furthermore, each runoff plot was investigated on two or three occasions to obtain a broader range of soil and surface properties, particularly a broader range in antecedent soil moisture content and vegetative cover. The research sites within the BV watershed were investigated on three occasions: spring (June), summer (August) and autumn (October) of 2012 for a total of 60 rainfall simulations. However, the sites within the STC watershed were only investigated on two occasions: spring (June – prior to crop emergence) and autumn (October – post-harvest) of 2013 as the tall and dense crops created unsuitable conditions for the rainfall simulation experiment during the summer growing season, for a total of 30 rainfall simulations. The runoff plots were not located in the exact same location between seasonal site visits

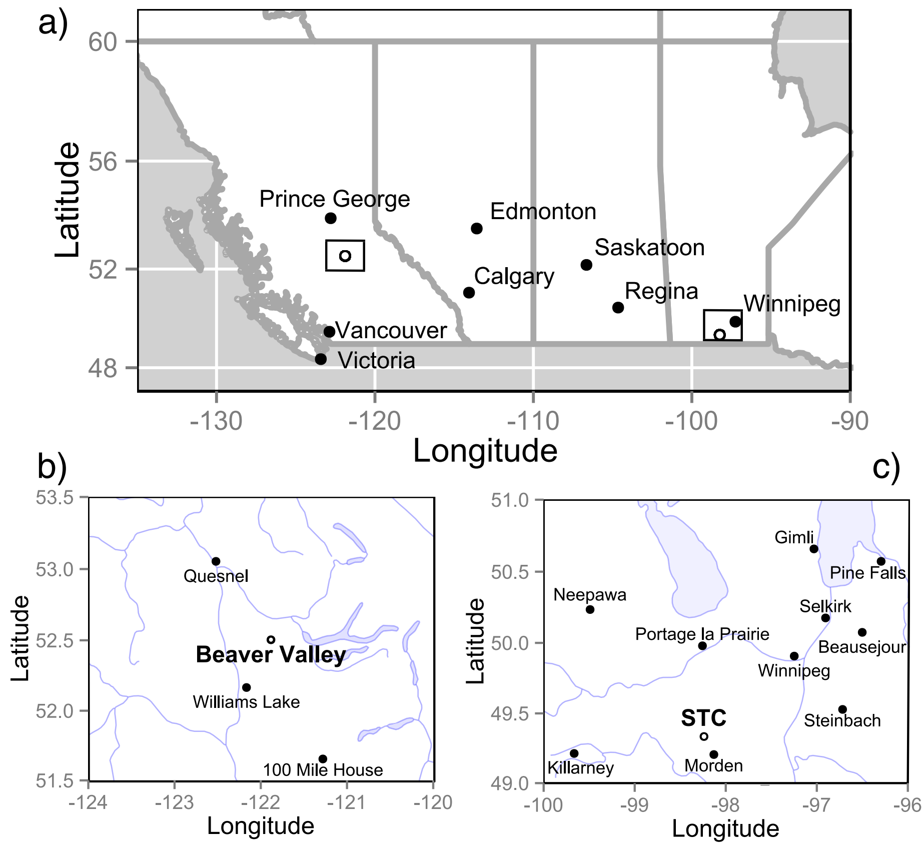

Fig. 1. Maps showing a) the provinces of south-western Canada and the locations (inset boxes) of the two sampling sites: b) Beaver Valley (BV) watershed and c) South Tobacco Creek (STC) watershed.

due to soil disturbance during the rainfall simulations, but all plots for a given transect and slope position were located within 2–3 m of each other. By locating the runoff plots on different hillslope positions, across different transects and in two distinctive agricultural locations, this rainfall simulator experiment reflects some of the spatial variability at the soil *catena*, landscape and regional scales. In addition, the measurements made during different seasons allowed for some of the temporal variability to be accounted for (e.g., vegetation cover, soil moisture, aggregate stability). For example, Stone and Walling (1997) found that the relative proportion of different grain-size fractions of sediment generated under simulated rainfall changed over the course of the year. Capturing this spatial and temporal variability allows generalizations to be developed about the significance of the various properties of the soil surface and their interactions in determining soil loss and the enrichment of fine-grained soil particles and SOC.

2.2. Rainfall simulator

A field-portable rainfall simulator, based on the design of Clarke and Walsh (2007), was used to generate runoff at the field sites described above. Briefly, this simulator requires no power, is gravity fed and is capable of producing rainfall intensities between 50 and 200 mm h^{-1} with a drop size distribution and kinetic energies similar to natural rainfall (Clarke and Walsh, 2007). For an intensity of 200 mm h^{-1} Clarke and Walsh (2007) reported a median drop size of 4.15 mm and a total storm kinetic energy of 727 J m^{-2} (over a 20-min storm event). Due to the small footprint of the rainfall simulator used in this study, the area of the runoff plots was limited to 0.16 m^2. The small runoff plot dimensions limited this study to the investigation of the selectivity of interrill or sheet erosion processes as the short slope length limited the formation of rills. The plot boundaries were constructed from stainless steel plates (40 cm long and 15 cm tall) and the plates were pushed 5–10 cm into the soil; care was taken to minimize soil disturbance and to ensure a water-tight seal with the soil during the installation process. On the down-slope edge a shallow triangular gutter collected the runoff into 1 L polyethylene bottles, similar to the design reported in Cao et al. (2015). Deionized water was used in the rainfall simulations to ensure consistency across all simulations. A rainfall intensity of 200 mm h^{-1} was used; while this intensity is high, it can provide information on the characteristics of the soil mobilized under rainfall and, importantly, within the requirements of this research, provide an adequate sample mass (>2 g) for analyses within a reasonable time frame. The rainfall simulation was run until there was 40 min of continuous runoff and the runoff was divided into two separate samples, again to ensure adequate sample mass for analysis. Within the literature, the reported time to reach a steady-state condition using rainfall simulators ranges from <5 min (e.g., Asadi et al., 2007) to >20 min (e.g., Armstrong et al., 2011) and this variability is related to differences in the applied rainfall rate and soil surface properties (e.g., texture, antecedent moisture, slope) (Proffitt et al., 1991). Within this study, the first 20 min of runoff (0–20 min) is likely more representative of easily erodible material while the second 20 min (20–40 min) is more representative of erosion under steady-state conditions. This sampling design is similar to the approach of Jin et al. (2009) where rainfall rates of 65–105 mm h^{-1} were applied over a 0.27 m^2 plot and runoff samples were collected in 15-min segments over a 90-min period. Due to low samples masses, Jin et al. (2009) aggregated the samples into two discreet samples (0–60 and 60–90 min) to allow for nutrient, SOC and particle size analysis. Despite the runoff sample aggregation Jin et al. (2009) found an effect of sampling period on the enrichment of SOC.

Source soil samples (0–2 cm) were collected immediately outside the plot boundaries prior to the start of the rainfall simulation in order to characterize the antecedent soil moisture content, SOC content and particle size distribution. The percent vegetative cover of the runoff plot was measured using digital photography and ImageJ image analysis software (Schneider et al., 2012). Photographs were taken with a Fujifilm FinePix Z33WP 10 M pixel camera held parallel to, and approximately 1.5 m from the surface. Each digital image covered the entire plot area (0.16 m^2) and was analyzed for vegetative cover by contrasting the difference in light-coloured vegetation with the dark-coloured soil (Koiter and Lobb, 2008). The gradient of the soil surface was measured using a clinometer (Jordán and Martínez-Zavala, 2008). A summary of the soil surface properties for both watersheds can be found in Table 1.

2.3. Laboratory analysis

2.3.1. Source soil and mobilized soil

A process flow diagram describing the sample preparation, subsampling and the analysis for both the source soil and mobilized soil is found in Fig. 2. The collected source soils were passed through a 2-mm sieve, the coarse stone fragments and large pieces of organic matter were removed and the sample was oven-dried. Prior to drying, a subsample was removed and stored at 4 °C and was used to measure the aggregated, or effective particle size distribution (EPSD) and for determination of the gravimetric moisture content. Runoff samples were left to settle in a cool dark room for 48 h and then the clear supernatant was siphoned off and the samples were oven-dried and weighed. Prior to settling, the runoff samples were gently stirred to resuspend the soil particles and a representative subsample was removed and stored at 4 °C and used for the determination of the EPSD.

2.3.2. Particle size analysis

The EPSD for the source soil samples was measured by combining field-moist soil with deionized water (1:20 ratio of soil to water) in a 25 mL centrifuge tube and placing the sample on a reciprocating shaker (25 rpm) for 10 min. The samples were then measured for EPSD immediately by laser diffraction (Malvern Mastersizer 3000, Malvern, UK). These samples were introduced directly to the dispersion unit (600 mL) and the pumping/stirring speed was set low (1800 rpm) to limit aggregate breakdown but fast enough to prevent settling. Similarly, the EPSD for the runoff samples was measured by introducing the sample directly into the dispersion unit with no prior disaggregation or removal of organic matter.

For the dispersed particle size distribution (DPSD), the oven-dried samples (up to 15 g) were weighed and placed in 100 mL beakers and deionized water was added to make a 1:5 ratio (soil to water) slurry. The samples were vigorously stirred for 15 min and allowed to settle for 12 h and the coarse organic matter that was floating on the surface was removed using a scoopula and was dried and weighed. The remaining sample was disaggregated using an ultrasonic probe (Misonix S-4000, Qsonica, Newtown, CT, USA) with a power output of 45 W and the duration of sonication for each sample was adjusted so that the amount of energy delivered was 300 J mL^{-1} (based on the total sample volume) (Yang et al., 2009). The samples were sonicated while in an ice bath and the sonication energy was pulsed (2 min on, 1 min off) to prevent the overheating of the sample and probe. The disaggregated samples were washed, using approximately 500 mL of deionized water, through nested 125- and 63-μm sieves resulting in three size fractions: 2000–125, 125–63 and <63 μm. The two larger particle size fractions were back-washed into glass beakers and the <63-μm fraction was transferred into a 1000 mL beaker and all three size fractions were oven-dried and weighed.

For the primary, or absolute particle size distribution (APSD), both source soil and mobilized soil samples received the same particle size analysis protocol that combines both sieving and laser diffraction techniques. For the <63-μm fraction, a subsample of both the oven-dried source and mobilized soil were digested with hydrogen peroxide (35%) to remove organic matter and an aliquot of sodium hexametaphosphate and sodium carbonate was added following the procedure of Kroetsch and Cang (2007). The sample was re-sonicated following the same procedure outlined above. The particle size of the <63-μm fraction was then measured using laser diffraction. The sieving and laser diffraction particle size techniques were merged by multiplying the percent <63 μm (by mass) by the percent in each size class measured by laser diffraction (by volume). This method assumes a uniform particle density and has been used by other researchers (e.g., Nadeu et al., 2011). The mass of the organic matter in each size fraction was accounted for based on the loss-on-ignition values.

Table 1
Summary of soil surface (0–2 cm) characteristics for both the Beaver Valley and South Tobacco Creek watersheds.

	Clay* (%)	Silt* (%)	Sand (%)	>2 mm (%)	Veg. cover (%)	Soil moisture (%)	Gradient (%)	Soil org. carbon (%)	Soil org. matter (%)	Particulate org. matter (%)	Runoff initiation (s)
Beaver Valley watershed											
Min.	4.5	22.4	17.6	2.1	4.1	3.2	1.7	2.8	6.4	3.3	30
1st Quartile	6.2	33.3	44.3	4.6	38.7	13.8	5.2	5.0	10.8	5.3	90
Median	8.4	42.9	47.5	7.7	68.9	31.9	8.7	6.3	12.9	7.2	135
Mean	8.6	41.3	50.1	12.3	62.8	35.2	8.9	6.1	12.9	10.2	189
3rd Quartile	10.0	46.1	60.3	16.2	86.7	50.7	10.5	6.8	14.1	12.7	240
Max.	17.4	67.2	72.8	40.6	100.0	110.7	21.3	9.6	19.5	33.8	660
South Tobacco Creek watershed											
Min.	10.9	47.0	9.3	0.0	2.0	12.7	1.7	1.2	3.6	0.7	30
1st Quartile	11.9	57.8	15.8	0.1	9.5	23.0	1.7	2.8	6.4	1.2	90
Median	14.7	61.3	24.0	3.3	20.3	31.3	5.2	3.5	8.4	1.5	120
Mean	15.7	61.1	23.3	5.2	34.0	32.0	11.7	3.3	8.4	2.1	193
3rd Quartile	19.7	65.0	31.1	7.9	51.0	38.2	14.5	4.2	10.7	1.9	300
Max.	22.6	75.4	34.9	24.4	98.0	61.8	51.0	5.1	12.8	17.4	600
Overall											
Min.	4.5	22.4	9.3	0.0	2.0	3.2	1.7	1.2	3.6	0.7	30
1st Quartile	7.2	39.9	27.8	3.5	19.5	20.6	5.2	3.9	9.1	2.0	90
Median	10.0	46.2	44.2	6.1	55.8	31.7	8.7	5.1	11.3	5.4	120
Mean	10.9	47.9	41.2	9.9	53.2	34.1	9.8	5.2	11.4	7.5	190
3rd Quartile	13.5	58.8	53.0	12.9	83.5	45.3	10.5	6.4	13.5	9.0	240
Max.	22.6	75.4	72.8	40.6	100.0	110.7	51.0	9.6	19.5	33.8	660

* Measured using laser diffraction which tends to underestimate clay and overestimate silt proportions relative to more common sedimentation techniques.

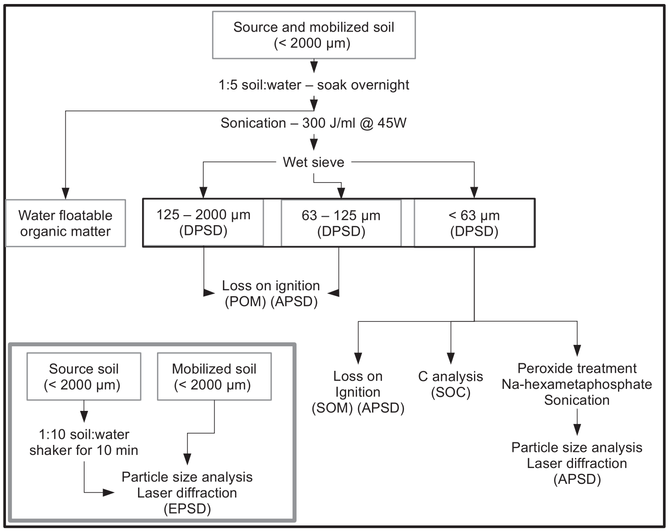

Fig. 2. Schematic showing the sample preparation, subsampling and analysis for the effective particle size distribution (EPSD; see inset), dispersed particle size distribution (DPSD), absolute particle size distribution (APSD), particulate organic matter (POM), soil organic matter (SOM) and soil organic carbon (SOC).

2.3.3. Soil organic carbon and organic matter analysis

Organic matter and SOC analyses followed the same protocol for both source and mobilized soil. Organic matter was determined for each of three particle size fractions (i.e., 2000–125, 125–63 and <63 μm) using the loss-on-ignition technique (550 °C for 1 h). The total mass of organic matter content was estimated by multiplying the total mass of each size fraction by the organic matter content (%). Particulate organic matter (POM) was estimated by summing of the organic matter content across the 2000–125 and 125–63 μm size fractions (Yang et al., 2009). Soil organic matter (SOM) was determined as the mass of organic matter in the <63-μm fraction (Yang et al., 2009).

Similar to the SOM analysis, SOC was measured as the C contained within the <63 μm size fraction; however, due to the large number of samples the particulate (>63 μm) organic carbon (POC) was not measured. No pretreatment was used to remove inorganic forms of carbon as test samples did not effervesce with the addition of 10% hydrochloric acid and the pH (1:2 ratio of soil to water) of the soils were <7.5 which indicate that carbonates are not present in significant quantities. Therefore, total C was used to estimate organic C content. Total C analysis was measured using an elemental analyzer (Elemental Combustion System ECS-4010, Costech Analytical Technologies Inc., Valencia, CA, USA). The SOC mass in each sample was estimated by multiplying the mass of the <63-μm fraction by the C content (%).

2.4. Data analysis

2.4.1. Enrichment ratios and aggregation index

Enrichment ratios (ER) are a convenient way to compare the properties of mobilized soil to the source soil. Enrichment ratio values >1 indicate an enrichment, and values <1 indicate a depletion as compared to the source soil. For example, the enrichment ratio for SOC (ER_{SOC}) for the mobilized soil was calculated as:

$$ER_{SOC} = [SOC]_{mobilized}/[SOC]_{source} \quad (1)$$

where $[SOC]_{\text{mobilized}}$ is the organic carbon content (%) in the mobilized soil and $[SOC]_{\text{source}}$ is the organic carbon content (%) in the source soil. Enrichment ratios for the different size classes and soil organic matter fractions were calculated using the same procedure.

The aggregate silt and clay (ASC) index (Igwe, 2000) was used to investigate the degree of aggregation in both the source and mobilized soil samples and was calculated as:

$$ASC = <63\mu m_{DPSD} - <63\mu m_{EPSD} \quad (2)$$

where <63 μm_{DPSD} is the sum of the clay and silt fraction (%) in the disaggregated form (DPSD) and <63 μm_{EPSD} is the sum of the clay and silt fraction (%) in the aggregated (EPSD) form.

2.4.2. Statistical analysis

All statistical analysis was undertaken using R Statistical Software v3.3.2 (R Core Team, 2016) through RStudio Integrated Development Environment v0.99.903 (RStudio, 2016). Linear mixed models (R package nlme version 3.1.128; Pinheiro et al., 2016) were used for all analyses with transect and runoff plot as random factors. Runoff plot was included as a random factor to control for the fact that the two runoff samples (i.e., 0–20 and 20–40 min runoff samples) are not independent (temporal pseudoreplication). Hillslope position (i.e., upper, mid and lower) was added as a covariate to control for any inherent variability along the length of a transect. Data were centred (i.e., predictors have mean of 0) prior to analysis to improve the interpretation of main effects and interactions. Since interaction between variables can influence the interpretation of main effects, all two-way interactions were investigated and added to the model in a backward stepwise process based on the likelihood ratio test (p-value < 0.1). Normality and homogeneity of variance were assessed visually and response variables were transformed, when necessary, to meet these assumptions. All models were checked for multiple collinearity (all variation inflation factors (VIF) were <5 and condition numbers (i.e., kappa) were <20) (Quinn and Keough, 2002).

All graphical plots were created using the R package ggplot2 v0.9.3.1 (Wickham, 2009). Differences in enrichment ratios between the two runoff periods were assessed by post-hoc pairwise comparisons with the Bonferroni p-value adjustment method when the model showed a significant main effect of runoff period or an interaction between the variable (e.g., size class) and the runoff period (R package multcomp version 1.4-0; Hothorn et al., 2008). Data were averaged across all runoff plots for each runoff period and plotted showing the standard error about the mean. For all other analyses, graphical plots were created with non-transformed data and show the model relation (regression line). Each relation was plotted while holding all other variables constant. In cases where interactions were significant the modelled relations represent ±1 standard deviation about the mean and data were also categorized based the mean value for a given soil surface property. Pearson correlation coefficients were calculated to assess the relation between enrichment ratios and soil loss as well as SOM and SOC content.

3. Results

3.1. Soil loss

There was a large range in the total mass of mobilized soil generated under the simulated rainfall. The distribution was heavily positively skewed and the soil loss was on average 37.5 (+100.1, −27.3) g m^{-2} per 20 min runoff period (±1 standard deviation, $\log_{10}$ back-transformed). There was a small difference in soil loss between the two runoff periods with the 0–20 min runoff period showing a higher soil loss when the antecedent soil moisture was high (Fig. 3; Table 2A). The soil loss was negatively correlated to the amount of vegetative cover (%) and showed a significant interaction with the antecedent soil moisture conditions and soil organic matter content. Low soil moisture conditions combined with low vegetative cover resulted in the higher soil loss and low soil organic matter content with low vegetative cover also resulting in higher soil loss (Fig. 3). Furthermore, runoff plots with a high slope gradient and a high soil moisture content tended

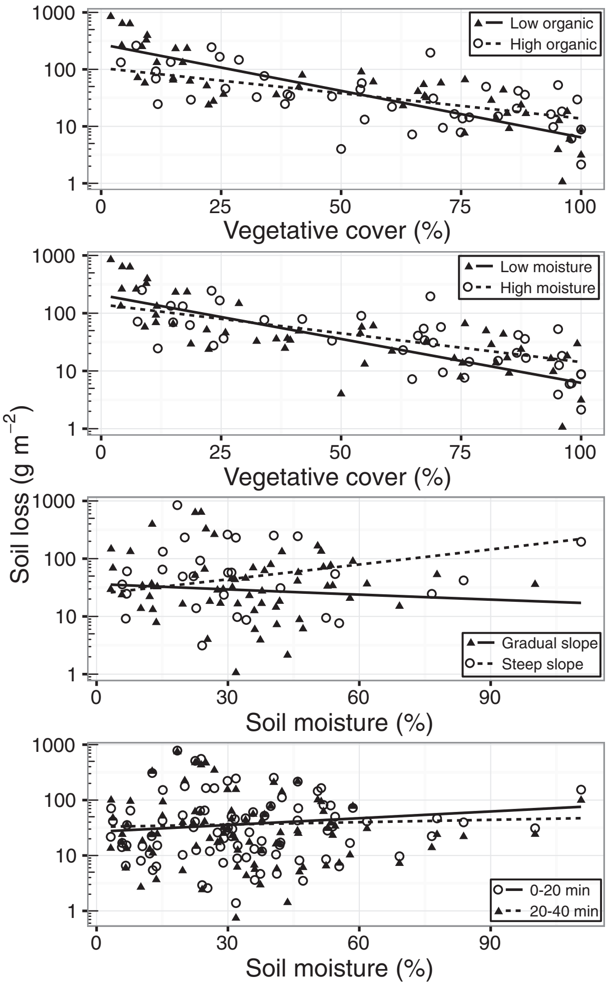

Fig. 3. The significant (p-value < 0.05) correlations between soil surface properties and soil loss. See Table 2(A) for additional details.

to have the greatest amount of soil loss and the effect of slope gradient diminished under drier soil surface conditions (Fig. 3).

3.2. Particle size selectivity

The runoff plots (i.e., source soil) investigated had average surface (0–2 cm) clay (<2 μm), silt (2–63 μm) and sand (63–2000 μm) contents of 10.9 ± 4.7%, 47.9 ± 13.3% and 41.2 ± 17.3%, respectively (±1 standard deviation). Overall, the mobilized soil generated under simulated rainfall were enriched in very fine-grained particles (all size fractions < 20 μm) and depleted in coarse-grained particles (all size fractions > 63 μm) as compared to the source soil for the APSD (Fig. 4a). The two smallest APSD fractions (<2 and 2–20 μm) had significantly lower ERs during the second runoff period as compared to the first runoff period. In contrast, the largest fractions (125–2000 μm) showed a significant increase during the second runoff period. Considering only the <63-μm fraction (Fig. 4b), there was still an enrichment of the very fine-grained particles (all size fractions <20 μm), but the values are closer to 1, and the difference between the two runoff periods is no longer significant. For the coarser silt fractions (20–40 μm and 40–63 μm), there was a depletion compared to the source soil and the ER values are significantly closer to 1 during the second runoff period.

The particle size enrichment ratios for the EPSD measurements (i.e., aggregate soil particles) show an enrichment in the three smallest particle size classes (<63 μm), a depletion in the largest size class (>125 μm) and the 63–125 μm fraction had values close to 1 (Fig. 5). In comparison with the APSD measurements (Fig. 4), the EPSD had ERs twice as high for the two finest fractions as well as enrichment ratios >1 for the size fractions between 20 and 125 μm whereas the APSD showed a depletion. Furthermore, there was higher variability in the ERs and there were no significant differences between the two runoff periods.

Similar to soil loss, the distribution of $ER_{<63}$ was heavily positively skewed and the ER was on average 1.41 (+0.38, −0.25) across both runoff periods (±1 standard deviation, inverse back-transformed). There was a small difference in $ER_{<63}$ (APSD) between the two runoff periods with the 0–20 min runoff period showing higher $ER_{<63}$ when the source soil had a low silt (%) content and this difference diminished as the silt content of the source soil increased (Fig. 6; Table 2B). Overall the $ER_{<63}$ was negatively correlated with the silt content (%) of the source soil. The effect of the gradient of the runoff plot on the $ER_{<63}$ showed an interaction with both the amount of vegetative cover and antecedent soil moisture content. The degree of enrichment was greater when the runoff plots had a gradual slope coupled with a higher moisture content and the effect of gradient diminished under dryer surface soil conditions (Fig. 6; Table 2B). Similarly, the degree of enrichment was smaller when the gradient of the runoff plot was higher and this effect diminished with increasing vegetative cover. Finally, the soil organic matter content of the surface soil had a small effect on the runoff plots with low soil organic matter resulting in a greater amount of enrichment. Overall, there was a negative correlation between $ER_{<63}$ and soil loss with a Pearson correlation coefficient of −0.34 (p-value < 0.001).

3.3. Organic matter selectivity

The source soils investigated had average surface (0–2 cm) SOC, POM and SOM contents of 5.2 ± 1.9%, 7.8 ± 7.3% and 11.4 ± 3.5%, respectively. Overall the mobilized soil generated under simulated rainfall were enriched in SOC, POM and SOM as compared to the source soil (Fig. 7a). Both the SOC and SOM enrichment ratios were significantly lower during the second runoff period (20–40 min). Considering only the <63-μm fraction (Fig. 7b) there is still an enrichment of the SOC and SOM, however, the values are closer to 1 and the difference between the two runoff periods are no longer significant. The enrichment of SOC (ER_{SOC}) was negatively correlated to the slope gradient (%), SOM (%) and <63-μm fraction (%) and positively correlated to soil moisture content (%) (Fig. 8; Table 2C). The SOM content of the source soil was the variable that has the largest effect on the ER_{SOC}.

There was a small difference in ER_{SOC} between the two runoff periods with the 0–20 min runoff period showing higher ER_{SOC} when the source soil had a low silt content and this difference diminished as the silt content of the source soil increased (Fig. 8; Table 2C). Overall the ER_{SOC} was negatively correlated with silt content of the source soil but showed an interaction with the soil organic matter content of the surface soil. Runoff plots with a lower soil organic matter content tended to result in the greatest amount of enrichment, but this effect diminished with increasing silt content. The antecedent soil moisture content was positively correlated with the ER_{SOC} when the slope gradient was gradual, however, under steeper slope gradients the antecedent moisture content had little effect on the ER_{SOC}. Similar to the $ER_{<63}$, there was a negative correlation between ER_{SOC} and soil loss with a Pearson correlation coefficient of −0.34 (p-value < 0.001).

3.4. The relation between SOC and particle size

The SOC content of both the source and mobilized soil was positively correlated with the ASC index (Fig. 9; Table 3A). There was not a significant interaction between the source and mobilized soil and the ASC index indicating that the relation was similar between the source and mobilized soil. Overall, there was a significant positive correlation of the SOC content and the amount of fine-grained particles (<63 μm) as well as a significant interaction between the amount of clay and silt and the source soil (Fig. 9; Table 3B). The relation between SOC and

Table 2
Results of linear mixed effects models looking at the correlation in soil loss (A), enrichment ratio (ER) of fine-grained sediment (B) and enrichment ratio (ER) of soil organic carbon (SOC) (C) with soil surface properties. Runoff period contrasts compares 0–20 min to 20–40 min.

Analysis	Parameter	Estimate	Std. Error	df	t value	*P*-value
	(Intercept)	1.662	0.065	85	25.538	<0.001***
	Vegetative cover (%)	−0.013	0.001	68	−10.295	<0.001***
	Gradient (%)	0.015	0.005	68	3.049	0.003**
	Silt (%)	0.006	0.003	68	1.763	0.082
	Soil organic matter (%)	−0.003	0.012	68	−0.247	0.806
	Soil moisture (%)	0.004	0.002	68	2.077	0.042*
(A) Soil loss	Runoff period	−0.003	0.023	85	−0.118	0.906
Log_{10} (g m^{-2})	Slope position (upper vs mid)	−0.172	0.086	68	−1.994	0.051
	Slope position (lower vs mid)	−0.133	0.084	68	−1.593	0.116
	Veg. cover (%) × Soil org. (%)	0.001	<0.001	68	2.790	0.007**
	Veg. cover (%) × Soil moist. (%)	<0.001	<0.001	68	2.136	0.036*
	Gradient (%) × Soil moisture (%)	0.001	<0.001	68	2.254	0.027*
	Gradient (%) × Runoff period	−0.004	0.003	85	−1.644	0.104
	Silt (%) × Soil org. (%)	0.002	0.001	68	1.981	0.052
	Soil moisture (%) × Runoff period	−0.003	0.001	85	−2.412	0.018*
	(Intercept)	0.677	0.012	86	58.538	<0.001***
	Vegetative cover (%)	−0.001	<0.001	69	−2.554	0.013*
	Gradient (%)	0.003	0.001	69	2.893	0.005**
	Silt (%)	0.011	0.001	69	16.487	<0.001***
	Soil organic matter (%)	0.006	0.002	69	2.495	0.015*
(B) Fine-grained soil (<63 µm) ER	Soil moisture (%)	−0.001	<0.001	69	−3.461	0.001**
$1/(<63_{runoff} / <63_{source})$	Runoff period	0.056	0.007	86	7.679	<0.001***
	Slope position (upper vs mid)	−0.011	0.014	69	−0.793	0.431
	Slope position (lower vs mid)	0.022	0.014	69	1.582	0.118
	Veg. cover (%) × Gradient (%)	<0.001	<0.001	69	−2.070	0.042*
	Silt (%) × Gradient (%)	<0.001	<0.001	69	−1.925	0.058
	Soil moisture (%) × Gradient (%)	<0.001	<0.001	69	4.556	<0.001***
	Silt (%) × Runoff period	−0.002	0.001	86	−4.022	<0.001***
	(Intercept)	1.690	0.084	85	20.012	<0.001***
	Vegetative cover (%)	<0.001	0.001	66	0.236	0.814
	Gradient (%)	−0.015	0.007	66	−2.071	0.042*
	Silt (%)	−0.023	0.004	66	−5.599	<0.001***
	Soil organic matter (%)	−0.050	0.013	66	−3.879	<0.001***
	Soil moisture (%)	0.003	0.002	66	1.554	0.125
(C) Soil organic carbon ER	Runoff period	−0.152	0.019	85	−7.927	<0.001***
$(SOC_{runoff} / SOC_{source})$	Slope position (upper vs mid)	0.044	0.064	66	0.691	0.492
	Slope position (lower vs mid)	−0.058	0.059	66	−0.980	0.331
	Soil org. (%) × Gradient (%)	−0.003	0.002	66	−1.964	0.054
	Soil moist. (%) × Gradient (%)	<0.001	<0.001	66	−2.027	0.047*
	Silt (%) × Soil moisture (%)	<0.001	<0.001	66	−1.766	0.082
	Silt (%) × Soil org. (%)	0.002	0.001	66	2.110	0.039*
	Silt (%) × Runoff period	0.007	0.001	85	4.954	<0.001***

* Significant at $p < 0.05$.
** Significant at $p < 0.01$.
*** Significant at $p < 0.001$.

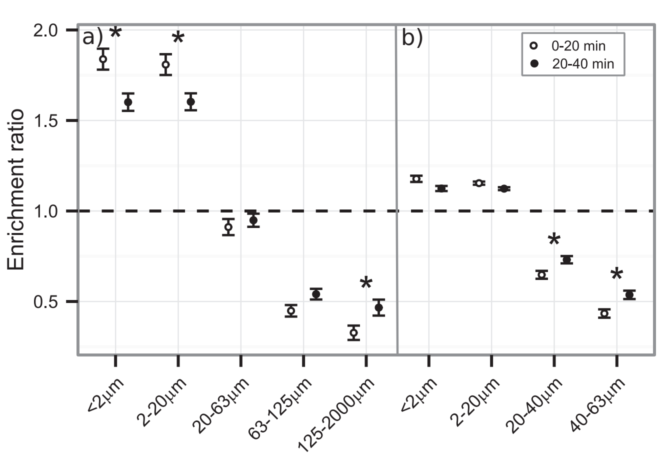

Fig. 4. Enrichment ratios for the eroded sediment with respect to the source soil for the different absolute particle size classes (mineral component only). Investigating the: a) total sediment (<2000 µm) and b) silt and clay only (<63 µm). Error bars indicate ±1 standard error and * denotes a significant difference (*p*-value < 0.05) between the 0–20 and 20–40 min runoff periods.

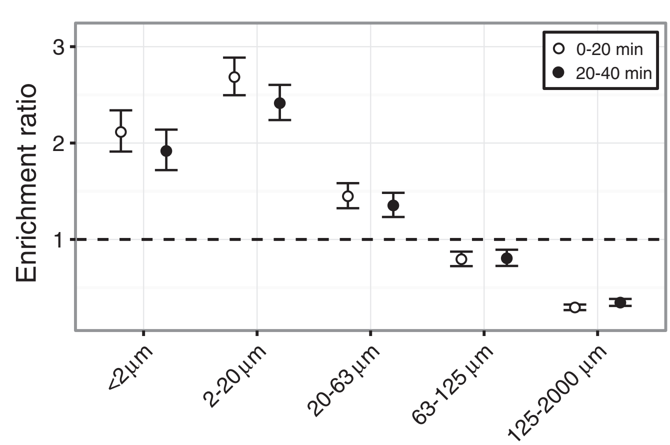

Fig. 5. Enrichment ratios for the eroded sediment with respect to the source soil for the different effective particle size classes (mineral and organic components; <2000 µm) Values have been log_{10} back-transformed. Error bars indicate ±1 standard error and * denotes a significant difference (*p*-value < 0.05) between the 0–20 and 20–40 min runoff periods.

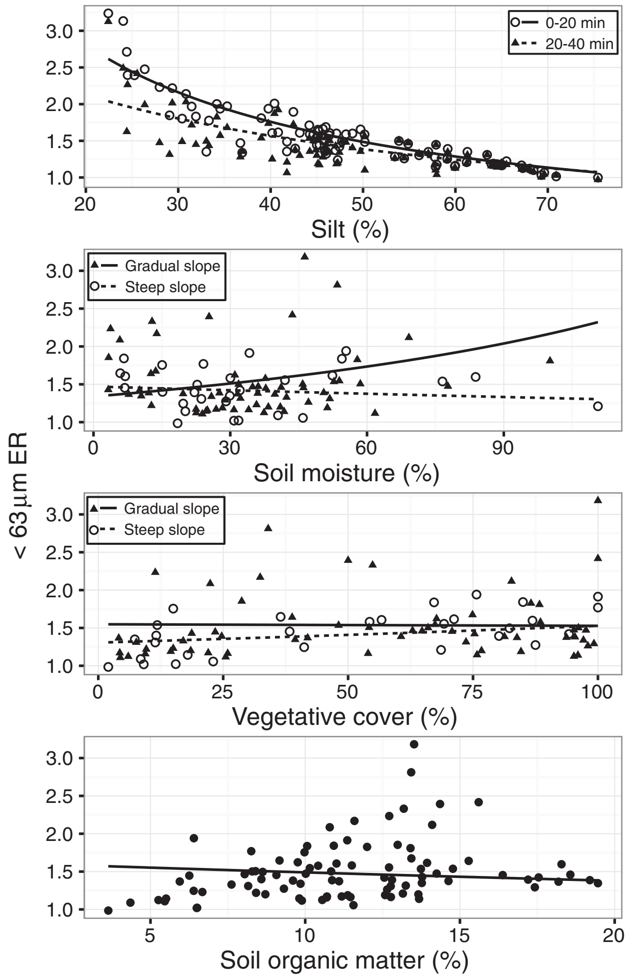

Fig. 6. The significant (p-value < 0.05) correlations between soil surface properties and the amount silt and clay (<63 μm; mineral component only) enrichment ratio of eroded sediment with respect to the source soils. See Table 2(B) for additional details.

the amount of fine-grained material for the source soil had a higher correlation compared to the mobilized soil and there was no difference between the two runoff periods. However, much of variation in the data, for both the ASC index and the clay and silt content, was explained by the random factor of transect and accounts for much of the spread of the raw data around the modelled relation.

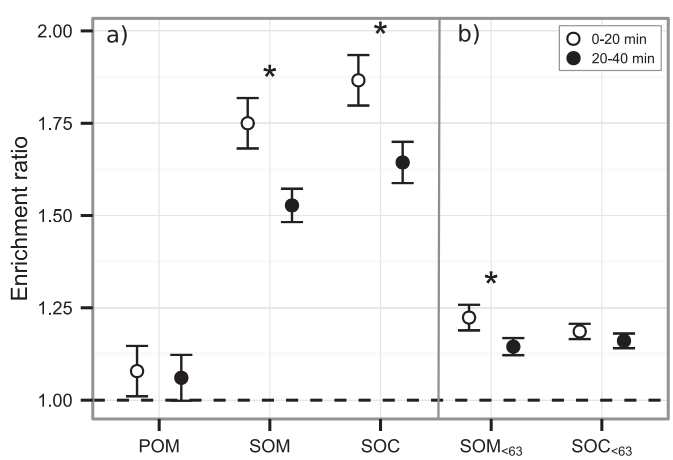

Fig. 7. Enrichment ratios for the eroded sediment with respect to the source soil for particulate organic matter (POM), soil organic matter (SOM) and soil organic carbon (SOC) fractions. Investigating the: a) total sediment (<2000 μm) and b) silt and clay only (<63 μm). Error bars indicate ±1 standard error and * denotes a significant difference (p-value < 0.05) between the 0–20 and 20–40 min runoff periods.

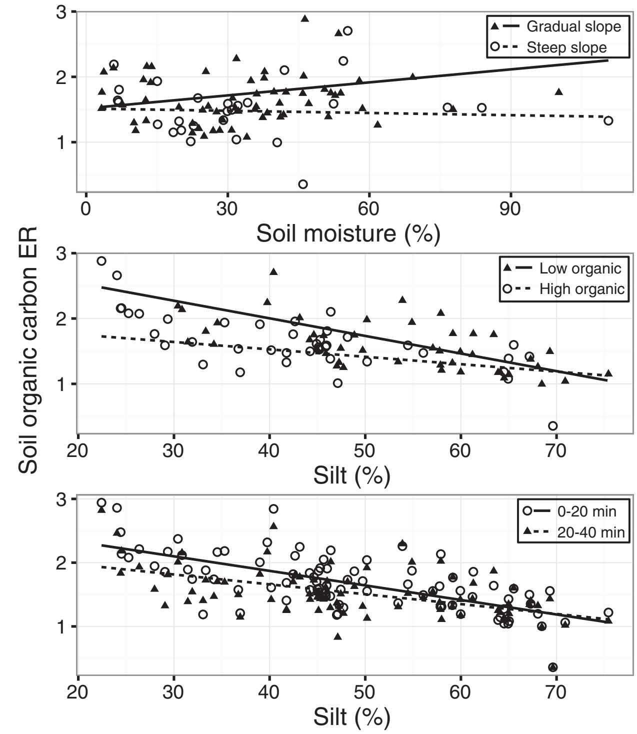

Fig. 8. The significant (p-value < 0.05) correlations between soil surface properties and the soil organic carbon enrichment ratio (ER) of eroded sediment with respect to the source soils. See Table 2(C) for additional details.

4. Discussion

4.1. Soil loss

The amount of soil loss as it relates to soil surface properties is generally consistent with the literature on interrill erosion. The inherent erodibility of the soil is primarily controlled by soil texture, SOM, soil structure, and permeability (Renard et al., 1991). Soils with a high silt and very fine sand content are typically the most erodible because this

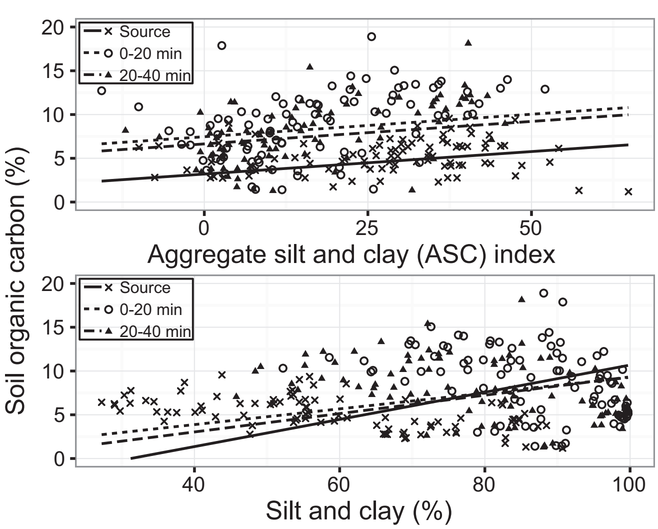

Fig. 9. The correlations between both the aggregated (ASC index) and absolute particle size (clay + silt) and the soil organic carbon content for the source soils and the eroded sediment. See Table 3 for additional details.

Table 3
Results of linear mixed effects models looking at the correlation in soil organic carbon with the aggregate silt and clay index (A), and soil organic carbon with the amount of clay and silt (B). Sample type contrasts compare Source vs 0–20 min (1) and 20–40 vs 0–20 min (2).

Analysis	Parameter	Estimate	Std. error	df	t value	*P*-value
(A) Soil organic carbon (%)	(Intercept)	8.224	0.814	172	10.102	<0.001***
	ASC index	0.051	0.007	172	7.516	<0.001***
	Source vs 0–20 min (1)	−4.264	0.204	172	−20.885	<0.001***
	20–40 vs 0–20 min (2)	−0.819	0.191	172	−4.289	<0.001***
	Slope position (upper vs mid)	0.120	0.308	77	0.391	0.697
	Slope position (lower vs mid)	0.717	0.314	77	2.287	0.025*
(B) Soil organic carbon (%)	(Intercept)	6.720	1.268	170	5.302	<0.001***
	Clay + silt (%)	0.090	0.012	170	7.473	<0.001***
	Source vs 0–20 min (1)	−0.247	0.255	170	−0.970	0.333
	20–40 vs 0–20 min (2)	−0.321	0.166	170	−1.929	0.055
	Slope position (upper vs mid)	−0.291	0.312	77	−0.932	0.354
	Slope position (lower vs mid)	1.137	0.319	77	3.561	<0.001***
	Clay + silt (%) × Sample type (1)	0.065	0.010	170	6.524	<0.001***
	Clay + silt (%) × Sample type (2)	0.015	0.010	170	1.504	0.135

* Significant at $p < 0.05$.
*** Significant at $p < 0.001$.

size class of soil particles does not form stable aggregates and as a result are easily mobilized and transported by overland flow (Shabani et al., 2014). Furthermore, silty soil tends to have lower infiltration rates as compared to soil with a higher percentage of sand which results in larger amounts of overland flow (Rawls et al., 1993). The results from this study are in agreement with this as there was a trend (p-value < 0.1) with an increase in the amount of soil loss with increasing silt content of the source soil (Table 2A).

Soil with a low SOM content also tended to be more erodible because SOM binds smaller soil particles together making them more resistant to mobilization and SOM improves infiltration and can reduce overland flow (Le Bissonnais and Arrouays, 1997; Franzluebbers, 2002). In this study, the effect of SOM on reducing soil loss was greatest when vegetative cover was low, and had a smaller effect under high vegetative cover conditions. The role of vegetative cover in this study is consistent with the literature on interrill erosion where the amount of vegetative cover is one of the most important factors influencing soil loss. For example, the STC watershed had an overall lower vegetative cover (34.0% ± 31.9) as compared to the BV watershed (62.5% ± 29.0) with a corresponding soil loss of >100% higher than the BV watershed. The amount of vegetative cover can change over time, and, as a consequence the potential for erosion is not constant throughout the year (Fig. 10). Therefore, during periods of low vegetative cover, SOM content has a greater contribution in reducing soil loss.

Dryer soil with low amounts of vegetative cover resulted in greater soil loss and this may be due to the breakdown of aggregates by rapid wetting (slaking and differential swelling) and through the impact of raindrops. The aggregate breakdown products result in smaller soil particles, which are more easily transported by erosional processes (Vermang et al., 2009). In contrast, low moisture soil with higher vegetative cover resulted in lower soil losses. The interception of raindrops by the vegetative cover reduces the amount of energy being transferred to the soil surface and the dry soil has a larger capacity to absorb water reducing the amount of overland flow. This is in contrast to the results of Li et al. (2015) where the antecedent moisture had a larger effect on soil loss as compared to vegetative cover with higher moisture soil generating higher soil losses. This contrast may reflect a difference in the relative contributions of both splash and interrill erosion. Splash erosion is more sensitive to changes in vegetative cover while interrill erosion is more affected by antecedent soil moisture content.

Antecedent soil moisture content can also impact soil erodibility through its influence on both infiltration and the stability of soil aggregates, as the rapid wetting of dry soil aggregates can result in disaggregation (i.e., slaking) (Warrington et al., 2009) which can then result in surface sealing reducing the infiltration rate and promoting overland flow (Sajjadi and Mahmoodabadi, 2015). For example, both Vermang et al. (2009) and Defersha and Melesse (2012) found that soil loss under simulated rainfall was greater for low moisture soil as compared to high moisture soil. The effect of soil moisture content prior to the rainfall simulation in this study was difficult to generalize, as there were significant interactions with the percent vegetative cover, slope gradient, and runoff period. There were negative Pearson correlation coefficients of −0.29 (p-value < 0.001) and −0.16 (p-value = 0.025) between the time since the beginning of the rainfall simulation and the initiation of runoff and both soil moisture and slope gradient, respectively. This suggests that, within this study, soil moisture may have a larger effect on the rate of runoff and this provides some evidence to explain the smaller effect of slope under lower soil moisture conditions.

The larger effect of slope gradient under wetter conditions may occur as a result of increased runoff due to saturation-excess overland flow. In general, wetter soil when rained upon will become saturated more quickly causing more rapid initiation of overland flow as compared to dryer soil. However, the soil moisture content throughout the soil profile needs to have been considered to fully assess the implications of saturation-excess overland flow on both the rate of runoff and

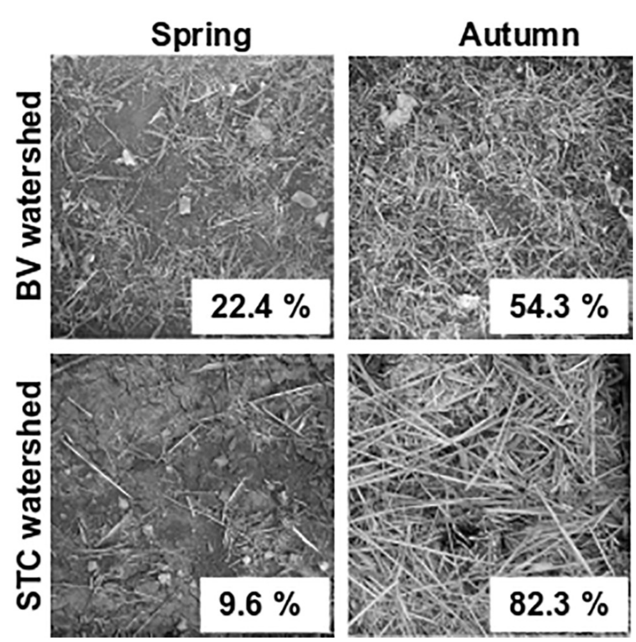

Fig. 10. An example of two sampling sites demonstrating the change in the percent vegetative cover between the spring and autumn seasons for the Beaver Valley (BV) and South Tobacco Creek (STC) watersheds with the percent cover in boxes.

soil loss. Interrill erosion is generally greater under steeper slope conditions as the gradient is proportional to the runoff velocity which increases the transport capacity of the runoff (Fox and Bryan, 1999). The overall effect of slope in this study was consistent with other research; steeper slope gradients had higher soil losses (e.g., Shi et al., 2012). Overall, the vegetative cover and the gradient of the runoff plot had the largest effect on soil loss, but the interactions with other soil surface properties demonstrate that soil loss due to interrill erosion is a complex process.

4.2. Particle size selectivity

It was expected that the larger particle size fractions (>63 μm; APSD) of the mobilized soil would be depleted with respect to the source soil as larger particles have a higher threshold shear stress and are less likely to be eroded (Fig. 4) (Shields, 1936). The response, in terms of the enrichment of the fine-grained fraction (<63 μm; APSD), was less predictable as this fraction has more potential to aggregate into larger particles that are more resistant to detachment and require greater amounts of energy to be transported as compared to the individual non-aggregated particles that comprise the aggregate soil particle. The factors that determine the differences in the particle size enrichment ratios between the EPSD and APSD (Figs. 4 and 5) are the disaggregation of aggregates during the simulated rainfall event and the removal of organic matter (POM and SOM) prior to the APSD measurement. It is difficult to separate the contribution of either component when comparing the two particle size measurement types. However, what is clear is that smaller particles were preferentially mobilized and transported as the higher enrichment ratios for the smaller particle size classes suggest that not enough energy, from raindrop splash or overland flow, was available to mobilize and transport the larger aggregates.

There are some interesting parallels in the results between the soil loss and the $ER_{<63}$ analysis (Figs. 3 and 6, respectively). Increases in either the slope gradient or the silt content of the source soil generally resulted in lower enrichment ratios and these same factors also resulted in higher soil losses. These properties, as discussed above, can result in an increased velocity and higher amounts of runoff, thus increasing the competence of the overland flow. These results are consistent with other research that has also shown that there is a significant correlation between soil loss or discharge and particle size selectivity (Armstrong et al., 2011; Yang et al., 2013). For source soil with a lower silt content, the 0–20 min runoff period had higher enrichment ratios compared to the 20–40 min runoff period and this may be due to the flush of small, loose and readily available soil particles at the soil surface. These results suggests that the supply of the most easily eroded size fraction is limited. This finding is consistent with Asadi et al. (2007) where the median particle size of the mobilized soil, generated under simulated rainfall, increased over time becoming closer in size to the source soil.

The interaction between soil moisture and the slope of the runoff plot on the $ER_{<63}$ is similar to soil loss where higher soil moisture and gradual slope gradient result in both lower soil loss and higher enrichment and this may be related to the lower transport capacity of the overland flow and reduced aggregate disintegration under these conditions. Overall, the amount of vegetative cover had a relatively small effect on the $ER_{<63}$ suggesting that raindrop impact on the soil surface had a small effect on the disintegration of soil aggregates and the resultant enrichment of fine-grained material. Both Shi et al. (2012) and Wang et al. (2014) found that soil mobilized under simulated rainfall had a higher enrichment of fine-grained particles with increasing mulching rates or lower kinetic energy.

The SOM content of a soil is generally positively correlated to the soil aggregate stability and size (Le Bissonnais and Arrouays, 1997). The increased stability of soil aggregates prevents the disintegration into smaller and more easily erodible particles and, therefore, limiting the loss of fine-grained soil particles (Leguédois and Bissonnais, 2004). Soil with a low degree of aggregation would experience less selectivity as a result of interrill erosion and have $ER_{<63}$ values closer to 1.0. It was unexpected that the SOM content of the runoff plot would have an overall negative correlation with the $ER_{<63}$. However, the variability between transects was very high and transects with higher SOM content had an overall higher $ER_{<63}$, but within a transect, plots with a higher SOM content had a lower $ER_{<63}$. This suggests that the role of SOM on the particle size selectivity is more site specific.

While the same soil surface properties that were significant factors in determining soil loss were also significant factors in the particle size selectivity, there were differences in their interactions. This indicates that there are some differences in the determining factors between soil loss and the particle size selectivity of interrill erosion processes.

The fact that soil erosion results in the preferential mobilization and transport of fine-grained soil particles is the one of the main reason that many sediment fingerprinting studies have sieved samples to <63 μm in an effort to limit the differences in particles size between source soil and sediment collected downstream. This allows for a more direct comparison in terms of biogeochemical properties (e.g., trace metal concentration, colour and cation exchange capacity) (e.g., Walling et al., 1999). The results from this study show that the sieving approach does reduce the differences in particle size between source and mobilized soil, however, it does not eliminate it all together. There is still an enrichment of the very fine-grained particles and a subsequent depletion of the coarse silt-size fraction after sieving. Furthermore, this study has also shown that the degree of selectivity is not static and can vary based on antecedent soil surface conditions and that there was a significant amount of spatial variability (i.e., the random factor transect was an important component of the overall variance). It is also important to put the relative significance of the selectivity of interrill erosion processes in perspective with other watershed processes (e.g., fluvial transport) in terms of determining the extent of downstream fining of sediment. For example, Koiter et al. (2015) found a reduction of ~55% in the median grain-size in less than 1 h of fluvial transport in an artificial channel.

The small enrichment of the very fine-grained particles after sieving still may present issues in the direct comparisons between mobilized and source soil as the smaller particles tend to have a higher specific surface area (SSA) and are more chemically reactive (Horowitz, 1991) and, therefore, geochemical and radiochemical elements of interest may be concentrated in these finer fractions. Researchers will sometimes use correction factors in addition to sieving procedures to account for the differences in particle size. A commonly used approach is to use the ratio of SSA between the sediment and source material and multiply it by the geochemical concentration of a given element (e.g., Collins et al., 1997). However, these correction factors can be problematic as it assumes that the relation between SSA and concentration is the same for all elements in terms of linearity, magnitude and direction, which is often not the case (Smith and Blake, 2014; Koiter et al., 2015).

4.3. Organic matter selectivity

The smaller enrichment ratio for the POM fraction as compared to the enrichment of SOM is likely due to the larger particle sizes (i.e., >63 μm) of this material which, as described earlier, requires more energy to mobilize and transport compared to smaller particles. However, for the mineral component of the mobilized soil there was a depletion in these particle size classes and the difference between the organic and mineral components in terms of enrichment is likely due to differences in particle density and shape. There is a large difference between the density of organic particles (0.9–1.3 Mg m^{-3}) and mineral particles (2.6–2.75 Mg m^{-3}) (Brady and Weil, 2001). Visual observations of the POM showed that most of the POM was either crop residues left after harvest (STC soil) or dead grasses (BV soil) and the particle shapes were typically blade shaped (long and flat) (Zingg shape classification; Zingg, 1935). Both the shape and density of the POM generally require less energy to mobilize compared to the mineral component of equivalent size (Le Roux, 2005). Both the SOM and SOC showed similar trends, as the two characteristics are highly correlated. The results show a higher degree of enrichment with the SOM as compared to the POM which is likely due to the smaller particle size of the SOM relative to the POM. Similar to the mineral fraction, sieving to <63 μm reduced, but did not eliminate, the difference between the source soil and the mobilized soil.

There are similarities between the $ER_{<63}$ and ER_{SOC} analysis in terms of some of the effects of the main factors and their interactions (Figs. 6 and 8). These similarities may, in part, be due to the SOC and the <63-μm size fraction aggregating together and being mobilized as aggregated soil particles. For example, the silt content and its interaction with the runoff period, as well as the interactions between the slope and antecedent soil moisture, have very similar effects on both the $ER_{<63}$ and ER_{SOC}. Furthermore, similar to the $ER_{<63}$ analysis, there was a significant negative correlation between soil loss and the ER_{SOC}, which is consistent with other research (e.g., Schiettecatte et al., 2008). Interestingly, the soil with the lower SOM, which also corresponds to soil with low SOC, had higher SOC enrichment ratios. This may be due to the lower aggregation potential of low SOM content soil and small non-aggregated and low density SOC-rich aggregate particles being more easily mobilized (Fig. 8). It is difficult to generalize this relation at a larger scale as the BV watershed soil had an average higher SOM content (12.9% ± 2.9) as compared to the STC watershed soil (8.4% ± 2.5) with a corresponding average ER_{SOC} of >20% higher than the STC watershed.

The enrichment of SOC or SOM can have similar effects as the enrichment of fine-grained soil particles in terms of the influence on geochemical composition of the material as organic matter also has a high SSA and is chemically reactive (Horowitz, 1991). There are also methods available to correct for differences in organic matter content and these are often applied in addition to particle size corrections. A commonly used approach is to use the ratio of SOM or SOC of the sediment to the source and multiply it by the geochemical concentration of a given element (e.g., Collins et al., 1997). However, these correction factors can be problematic as they assume that the relations between SOM or SOC and concentration are the same for all elements in terms of linearity, magnitude and direction. Furthermore, the use of a particle size correction factor in addition to an organic matter correction factor also assumes that the effects of organic matter and particles size are additive, but the amount of fine-grained particles and organic matter are often positively correlated (Ma et al., 2015). Further research is clearly needed to assess these correction factors. Consequently, several researchers have identified that correcting for both particle size and organic matter selectivity should be treated with caution and potentially avoided (Smith and Blake, 2014). Furthermore, the organic matter selectivity of the water erosion process has implications for the redistribution of C within the landscape and its loss to surface waterways. When assessing the role of soil erosion in C dynamics this research shows that there is a disproportional amount of C being mobilized and transported relative to the source soil (i.e., ER_{SOC} is typically >1) and that total soil loss is insufficient in characterizing C redistribution and loss.

4.4. The relation between APSD, EPSD and SOC

The results for this study show that SOC is positively related to the degree of aggregation in soil, which is consistent with the concept of SOC and its associated microorganisms acting as binding agents. The mobilized soil had an overall higher SOC content, but a similar relation with the aggregation index, suggesting that SOC-rich aggregates are being preferentially mobilized and transported and are stable (i.e., do not breakdown due to the simulated rainfall). This phenomenon may be related to the differences in densities between SOM and the mineral component. In addition, Williams and Petticrew (2009) found that the SOC content was an important factor influencing aggregate stability. The amount of fine-grained soil particles was positively correlated to the ASC index (data not shown) in source soil as clay. Similar SOC can act to bind particles together. As demonstrated in this study, the role of both fine-grained particles and SOC in promoting aggregation are consistent with other studies (e.g., Ma et al., 2014).

The amount of fine-grained soil particles mobilized was positively correlated to SOC content in both the source and mobilized soil, which is consistent with other research (Feller and Beare, 1997). This correlation may be due to the long-term higher nutrient and moisture content often associated with fine-textured soil which promotes vegetation growth. Increasing amounts of clay and silt can promote aggregation and the formation of organo-mineral complexes which can protect SOC from decomposition. The steeper regression line for the source soil, as compared with the mobilized soil, is likely due to the preferential mobilization and transport of fine-grained soil particles as the presence of sand in the source soil acts to dilute the SOC measurements (e.g., mass and density differences). The effect of sand-sized soil particles on SOC can also be seen by comparing Fig. 7a and b whereby the removal of sand size particles reduces the difference (i.e., ER closer to 1) between source and mobilized soil. The high variance component associated with the random factor transect suggests that the exact relation between SOC and measures of particle size (i.e., APSD and EPSD) is site-specific.

5. Conclusions

Soil mobilized under simulated rainfall resulted in an enrichment of both fine-grained soil particles and SOC relative to the source soil. There was an enrichment of both aggregated and dispersed soil particles <20 μm with the aggregated particles having a greater amount of enrichment. This demonstrates that the EPSD was more affected by interrill erosion processes than the APSD. Furthermore, the ERs of SOM, SOC and particle sizes <20 μm (APSD) were greater for the 0–20 min as compared to the 20–40 min runoff period providing evidence that there is a flush of easily erodible small and organic-rich soil particles. This may also provide an indication that there is a change in the erosional process (splash vs. overland flow) between the two sampling periods. Furthermore, the mobilized soil had an overall higher SOC content, but a similar relation with the aggregate silt and clay index, providing evidence that the SOC-rich aggregates being preferentially mobilized have a high degree of stability. Sieving to <63 μm, a common approach to limit the differences in particle size and SOC between source and eroded soil, did reduce the enrichment ratio but did not eliminate it. This demonstrates that further procedural steps, including sieving to a smaller particle size may be needed to account for these differences which will allow a more direct comparison of biogeochemical properties between source and mobilized soil.

There were similarities in terms of soil surface properties and their interactions and the subsequent effects on soil loss and the selectivity of small and organic-rich particles. The soil surface moisture content and its interaction with the gradient of the runoff plot were common across all three analysis where high soil moisture coupled with gradual slope resulted in lower rates of soil loss and greater enrichment of fine-grained material and SOC. There were also some differences in the main factors determining the soil loss including the amount of vegetative cover (%); while this had a large effect on soil loss it had a relatively small effect on the $ER_{<63}$ and no effect on the ER_{soc}. The enrichment of both fine-grained soil particles and SOC were negatively correlated with soil loss providing evidence that the rate of soil loss can be used to estimate the degree of selectivity. However, the differences between the factors and their interactions that determine soil loss and selectivity demonstrate that predictions of particle size and SOC enrichment based on the rate soil loss alone may not adequately describe the process of selectivity. Overall, it was difficult to rank the relative importance of the individual soil surface properties that influence particle size and organic matter selectivity as there were significant interactions between some of the soil surface properties.

This study investigated the selectivity of interrill erosional processes on the fine-grained soil particles and SOC under simulated rainfall conditions. However, this research has also only considered interrill erosion by raindrop splash and overland flow over a short distance and the assessment of other erosional processes (e.g., rill, gully, wind and tillage erosion) and their interactions on selectivity are an important research objective. Soil erosion is just the first step in the sediment cascade and there will be additional selectivity in each step including the delivery through riparian zones (Syversen and Borch, 2005) and fluvial transport downstream (Koiter et al., 2015). There is an important research need to link each of these steps in terms of the magnitude and the determining factors of the particle size and SOC enrichment (Koiter et al., 2013). This will allow for the improvement of process-based watershed models and management tools.

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