Dryland, Calcareous Soils Store (and Lose) More Near-Surface Organic Carbon Than Previously Thought

Authors:

Andrew M. Cunliffe[1]*, Alan K. Puttock[1], Laura Turnbull[2], John Wainwright[2] and Richard, E. Brazier[1]

Author Institutions:

[1]University of Exeter, [2]Durham University

[*]Corresponding Author

Keywords:

Soil Organic Carbon (SOC); Semi-Arid; Enrichment; Stabilization Mechanism; Erosion; Shrub encroachment

Key points:

Stable coarse (>2 mm) aggregates can contain significant proportions of total soil organic carbon

Calcium carbonate precipitation may stabilize organic carbon in dryland soils

Erosion-induced organic carbon yields are higher from shrublands compared to grasslands

Abstract

Semi-arid ecosystems are susceptible to changes in dominant vegetation which may have significant implications for terrestrial carbon dynamics. The present study examines the distribution of organic carbon (OC) between particle size fractions in near-surface (0-0.05 m) soil and the water erosion-induced redistribution of particle-associated OC over a grass-shrub ecotone, in a semi-arid landscape, subject to land degradation. Coarse (>2 mm) particles have comparable average OC concentrations to the fine (<2 mm) particles, accounting for ~24-38% of the OC stock in the near-surface soil. This may be due to aggregate stabilization by precipitated calcium carbonate in these calcareous arid soils. Critically, standard protocols assuming coarse fraction particles contain no OC are likely to underestimate soil OC stocks substantially, especially in soils with strongly stabilised aggregates. Sediment eroded from four hillslope scale (10 x 30 m) sites during rainstorm events was monitored over four annual monsoon seasons. Eroded sediment was significantly enriched in OC; enrichment increased significantly across the grass-shrub ecotone, and appears to be an enduring phenomenon probably sustained through the dynamic replacement of preferentially removed organic matter (OM). The average erosion-induced OC event yield increased six-fold across the ecotone from grass-dominated to shrub-dominated ecosystems, due to both greater erosion and greater OC enrichment. This erosional pathway is rarely considered when comparing the carbon budgets of grasslands and shrublands, yet this accelerated efflux of OC may be important for long-term carbon storage potentials of dryland ecosystems.

1. Introduction

Drylands are extensive ecosystems, covering around 40% of the land surface and directly providing ecosystem services to some 2.4 billion people [*Adeel et al.*, 2005; *Reynolds et al.*, 2007]. Although dryland soils usually contain only small amounts of organic carbon (OC) per-unit-area, their extent and low turnover rates means they contain an estimated 10-27% of the OC stock in terrestrial soils [*Safriel et al.*, 2005; *Finch*, 2012]. Recently it has also been argued that dryland ecosystems may contribute significantly to interannual variations in the global carbon cycle [*Poulter et al.*, 2014].

Critically, dryland ecosystems are susceptible to a range of degradation processes such as wild fire, and the erosion of soil and soil-associated nutrients by overland flow during infrequent but high-intensity rainstorm events [*Adeel et al.*, 2005; *Maestre et al.*, 2006; *Turnbull et al.*, 2010b, 2011; *Wainwright and Bracken*, 2011; *Michaelides et al.*, 2012; *Bestelmeyer et al.*, 2015]. One of the greatest uncertainties in our understanding of carbon dynamics in drylands is associated with degradation which can reduce carbon storage in both biomass and soil stock by: (i) combusting organic matter [*Sankey et al.*, 2012; *Poulter et al.*, 2014; *Ahlström et al.*, 2015], (ii) decreasing photosynthetic uptake by vegetation [*Lal*, 2001], (iii) accelerating decomposition and photodegradation of organic matter [*Foereid et al.*, 2011; *Barnes et al.*, 2012], and (iv) accelerating erosional losses to fluvial systems [*Lal*, 2001, 2003; *Brazier et al.*, 2013; *Puttock et al.*, 2013, 2014]. Globally, the degradation of dryland ecosystem carbon storage capacity is estimated to release ~0.3 Pg C yr^{-1} to the atmosphere from terrestrial stocks [*Adeel et al.*, 2005; *Safriel et al.*, 2005], and significantly influences the global biogeochemical carbon cycle [*Schlesinger et al.*, 1990; *Qi et al.*, 2001; *Poulter et al.*, 2014]. However, there is large uncertainty regarding the fate of eroded OC, some of which is released to the atmosphere [*Van Oost et al.*, 2005; *Lal and Pimentel*, 2008].

The encroachment of woody shrubs into grasslands is a widespread phenomenon globally [*Van Auken*, 2009; *Eldridge et al.*, 2011]. This change in plant functional type, amongst other things, alters ecosystem carbon dynamics with potentially significant implications for global biogeochemical carbon cycling [*Schlesinger et al.*, 1990; *Pacala et al.*, 2007; *Barger et al.*, 2011]. While much work has been undertaken to characterise carbon stocks in semi-arid grasslands and shrublands, the net carbon effect of the vegetation transitions varies with environmental context [*Conant et al.*, 1998; *Jackson et al.*, 2002; *Barger et al.*, 2011] and significant uncertainty remains regarding the controls on the various carbon fluxes and pools in these ecosystems [*Goodale and Davidson*, 2002; *Jackson et al.*, 2002; *Pacala et al.*, 2007]. Comparisons of the carbon budgets of grasslands and shrublands usually assume that the lateral redistribution of carbon is insignificant [e.g., *Petrie et al.*, 2015]; however, it is well established that changes in ecosystem structure following shrub encroachment into semi-arid grasslands can accelerate the erosion of soil and soil-associated chemicals [*Schlesinger et al.*, 2000; *Wainwright et al.*, 2000; *Ridolfi et al.*, 2008; *Turnbull et al.*, 2010b, 2011; *Brazier et al.*, 2013; *Puttock et al.*, 2013, 2014]. Recent work has indicated that the erosion-induced efflux of carbon from semi-arid shrublands may be substantially higher than that from comparable grasslands [*Brazier et al.*, 2013; *Puttock et al.*, 2013], and that this flux includes the loss of previously stable legacy carbon [*Puttock et al.*, 2014]. Therefore, to constrain understanding of the impact of shrub encroachment on the carbon dynamics in semi-arid rangelands, the aim of this study is to examine the water erosion-induced redistribution of particle-associated OC at different sites across a semiarid grass-shrub ecotone.

Most knowledge of soil organic carbon (SOC) dynamics as impacted by erosion originates from studies in intensively-managed agroecosystems dominated by tillage erosion, often in temperate regions [e.g., *Lal*, 2005; *Beniston et al.*, 2015; *Lacoste et al.*, 2015]. However, as several workers have noted, process understanding obtained from this work is not always directly transferable to less intensively managed ecosystems, in other environmental contexts [*Parsons et al.*, 1991; *Bryan*, 2000; *Mayeux*, 2001; *Liao et al.*, 2006b]. Therefore, it is important to extend detailed monitoring to unmanaged natural ecosystems, to evaluate transferability of existing process understanding.

Relative to contributing topsoils, eroded sediments are commonly enriched in particle-associated chemicals, such as OC [*Jacinthe et al.*, 2001; *Lal*, 2003, 2005; *Lal et al.*, 2004]. OC enrichment has been observed in laboratory simulations [*Sharpley*, 1985; *Palis et al.*, 1997; *Polyakov and Lal*, 2004b; *Kuhn*, 2007; *Jin et al.*, 2009; *Hu et al.*, 2013], interrill erosion plots [*Lal*, 1976; *Cogle et al.*, 2002; *Jin et al.*, 2008; *Brazier et al.*, 2013; *Puttock et al.*, 2013; *Wang et al.*, 2013b, 2014a] and at catchment-scales [*Starr et al.*, 2000; *Owens et al.*, 2002; *Rhoton et al.*, 2006; *Wang et al.*, 2010; *Nadeu et al.*, 2011, 2012; *Meixner et al.*, 2012], and is significant, because it precludes the accurate estimation of chemical fluxes on the basis of mass of sediment eroded and chemical concentration in the contributing soil. Organic carbon (OC) is typically associated with finer and less dense particles so OC enrichment is thought to depend on the selectivity of the dominant detachment, transport and deposition processes, which varies both spatially and temporally [*Owens et al.*, 2002; *Jacinthe et al.*, 2004; *Schiettecatte et al.*, 2008a; *Jin et al.*, 2008, 2009; *Turnbull et al.*, 2010b; *Nadeu et al.*, 2011, 2012; *Hu et al.*, 2013; *Wang et al.*, 2014a]. For example, OC enrichment is thought to decrease during higher intensity and larger magnitude rainstorms, as the dominance of highly selective interrill erosion processes is exceeded by less-selective rill erosion processes [*Schiettecatte et al.*, 2008a; *Wang et al.*, 2014a].

Several workers have argued that OC enrichment is not significant as a long-term, large-scale phenomenon, on the basis that: (i) OC enrichment is thought to decrease over increasing spatial scales as the dominance of highly selective interrill erosion processes is surpassed by less selective concentrated flow erosion [*Schiettecatte et al.*, 2008a; *Van Oost et al.*, 2008],

(ii) the OC mass balance in the contributing soil is preserved [*Kuhn and Armstrong*, 2012; *Hu et al.*, 2013], and (iii) sedimentary deposits in lakes and reservoirs often contain OC concentrations near-parity with the contributing topsoils [*Ritchie*, 1989; *Stallard*, 1998]. However, these contentions are challenged by the knowledge that: (i) rill erosion processes often exhibit at least some selectivity for particle size and density [*Parsons et al.*, 1991, 1994; *Malam Issa et al.*, 2006] and enrichment is observed at catchment-scales [*Starr et al.*, 2000; *Owens et al.*, 2002; *Rhoton et al.*, 2006; *Wang et al.*, 2010; *Nadeu et al.*, 2011, 2012; *Meixner et al.*, 2012]. (ii) The dynamic replacement of organic matter (OM) inputs to the soil surface [*Harden et al.*, 1999; *Li et al.*, 2007; *Berhe et al.*, 2008; *Doetterl et al.*, 2012] could sustain preferential removal of particle-associated OC without depleting the contributing soil, preserving the mass balance. (iii) Without enrichment, deposited sediments should exhibit depletion in OC concentrations relative to the eroding soil. This is because carbon-rich particles are less likely to be deposited due to relatively low densities and small sizes [*Starr et al.*, 2000; *Jacinthe and Lal*, 2001; *Lal*, 2003, 2005; *Beuselinck et al.*, 2000; *Schiettecatte et al.*, 2008b; *Nadeu et al.*, 2011, 2012], and more suggestions that the decomposition of mobilized OC is accelerated due to both aggregate disruption during erosion and transport reducing physical protection [*Polyakov and Lal*, 2004b; *Lal et al.*, 2004; *Lal*, 2005; *Mora et al.*, 2007; *Schiettecatte et al.*, 2008a; *Jin et al.*, 2009] and also priming effects due to combining labile and recalcitrant OC [*Kuzyakov*, 2010; *Bianchi*, 2011].

Numerical modelling approaches are a valuable tool to understanding the erosion-induced redistribution of OC over large spatial and temporal scales [*Polyakov and Lal*, 2004a; *Schiettecatte et al.*, 2008a; *Quinton et al.*, 2014]. However, the belief that OC enrichment was insignificant led to numerical model development which either ignored the process of OC enrichment [e.g., *Voroney et al.*, 1981; *Mitchell et al.*, 1998; *Fierer and Gabet*, 2002; *Quinton et al.*, 2014], or which represented it via a single, poorly validated coefficient [e.g., *Bouwman*, 1989; *Lee et al.*, 1996; *Starr et al.*, 2001]. Clearly, there is a need to improve process representation of OC redistribution in numerical models, but most information on the mechanisms of OC enrichment originates from highly reductionist experiments, often using small plots of homogenised repacked soils with synthetic structure, subjected to artificial rainfall [e.g., *Ghadiri and Rose*, 1991b, 1991a; *Palis et al.*, 1990b, 1990a; *Proffitt and Rose*, 1991; *Wan and El-Swaify*, 1997, 1998; *Kuhn*, 2007; *Schiettecatte et al.*, 2008a; *Jin et al.*, 2009; *Hu et al.*, 2013; *Hu and Kuhn*, 2014]. Consequently, there are large uncertainties regarding the transferability of knowledge to the redistribution of soil-associated OC in natural ecosystems subject to natural rainfall events [*Glenn et al.*, 1998; *Lal et al.*, 2001; *Polyakov and Lal*, 2004a; *Kuhn*, 2007; *Nadeu et al.*, 2011, 2012; *Doetterl et al.*, 2012]. Although many studies have attributed OC enrichment predominantly to the preferential erosion of fine, OC-rich particles [e.g., *Nelson et al.*, 1994; *Balesdent et al.*, 1998; *Guibert et al.*, 1999; *Rhoton et al.*, 2006; *Wang et al.*, 2013a], recent work has suggested that the enrichment of fine particles alone cannot explain observed OC enrichment [Z. *Wang et al.*, 2010, 2013b; *Chartier et al.*, 2013].

Standard protocols for measuring soil organic carbon (SOC) discard the coarse (>2 mm) particle size fraction, assuming it contains no OC [*Robertson and Paul*, 2000; *Lal and Kimble*, 2001; *Ellert et al.*, 2001; *Bird et al.*, 2002; *Jackson et al.*, 2002; *Ewing et al.*, 2007; *Throop et al.*, 2012; *Sankey et al.*, 2012; *Frank et al.*, 2012; *Brazier et al.*, 2013; *Puttock et al.*, 2013, 2014]. However, work in a variety of environmental contexts has demonstrated that coarse (>2 mm) particles can contain OC concentrations comparable to the fine (<2 mm) fraction, accounting for 5% of the total SOC stock [*Corti et al.*, 2002; *Agnelli et al.*, 2000, 2002]. In calcareous dryland soils, the precipitation of calcium carbonate can stabilize macroaggregates [*Bryan*, 2000; *Nash and McLaren*, 2003; *Alonso-Zarza and Wright*, 2010]. Such stabilized aggregates may incorporate OC associated with fine particles, or fine particulate organic matter (POM) [*Duchaufour*, 1976; *Goudie*, 1996; *Baldock and Skjemstad*, 2000], particularly as the biochemical actions of roots and fungi facilitate calcium carbonate precipitation in arid soils [*Goudie*, 1996; *Alonso-Zarza and Wright*, 2010; *Gocke et al.*, 2011]. Therefore, the OC concentration of coarse (>2 mm) particles needs to be examined to assess whether there may be underestimation of SOC inventories in calcareous dryland soils.

In summary, this study has four objectives: (i) to examine potential OC storage in coarse (>2 mm) particles in calcareous soils. (ii) To determine whether there are systematic changes in the enrichment of OC across an ecotone of changing plant functional types from a grass-dominated to a shrub-dominated ecosystem. (iii) To investigate controls on OC enrichment in natural ecosystems subjected to natural rainfall events, quantifying the extent to which particle size selectivity can explain observed OC enrichment. (iv) To quantify differences in erosion-induced effluxes of OC across an ecotone from a grass-dominated to a shrub-dominated ecosystem over a four-year period.

2. Methods

2.1. Study site

The study site is located in the Mackenzie Flats of the Sevilleta National Wildlife Refuge (SNWR) in central New Mexico, USA (34°19’N, 106°42’W), experiencing a semi-arid climate with 256 mm mean annual precipitation of which ~60% falls during the summer monsoon period. Soil series are shallow and classified as Turney loams overlaying a well-developed petrocalcic horizon located ~0.25-0.45 m below the surface [*Kieft et al.*, 1998; *Rawling*, 2005; *Turnbull et al.*, 2008b].

2.2. Experimental Design and Sampling

Four, 300-m^2 (30 m × 10 m) experimental sites were examined, across a grass-shrub ecotone from black grama (*Bouteloua eriopoda*) dominated communities to creosotebush (*Larrea tridentata*) dominated communities. These sites were selected to examine interactions between surface vegetation cover and ecosystem functioning, so were selected to be topographically similar, with relatively planar slopes. Previous work at these sites across this grass-shrub ecotone has examined differences in abiotic and biotic ecosystem structure [*Turnbull et al.*, 2010a], hydrology and sediment dynamics [*Turnbull et al.*, 2010b, 2010c], hydrological connectivity [*Puttock et al.*, 2013], nitrogen and phosphorus dynamics [*Turnbull et al.*, 2011] and organic carbon dynamics [*Puttock et al.*, 2012, 2014; *Brazier et al.*, 2013]. Within each site, five 236 cm^3 samples of near-surface soil were collected from random locations beneath each surface cover (bare soil and, where present, grass and shrub), totalling 10-15 samples per site. Samples were collected by driving a ring sampler (0.0775 m diameter, 0.05 m depth) into the soil. The surrounding soil was excavated from around the sampler, and a pointing trowel was used to slice the sampler out of the soil so that the soil surface was flush with the sampler [*Brazier et al.*, 2013]. Samples were analysed separately for bulk density, particle size distribution (PSD) and OC concentration. The 0-0.05 m soil sampling depth was selected because this near-surface layer is highly susceptible to interaction with surface-transport processes at hillslope scales, in accordance with similar research undertaken in these environments [e.g., *Wainwright et al.*, 2000; *Rhoton et al.*, 2006; *Li et al.*, 2007; *Turnbull et al.*, 2010b, 2010a; *Puttock et al.*, 2012, 2014; *Brazier et al.*, 2013]. 37 discrete rainstorm events were monitored over the four sites in the four summer monsoon periods, covering both wetter- and drier-than-average monsoon seasons [*Petrie et al.*, 2014]. Precipitation and runoff were monitored at one-minute resolution. Overland flow and associated eroded sediment was captured in stock tacks, which contained all runoff and sediment in 84% of events, with the six occurrences of tank exceedance distributed across all plots. This total capture is important because partial sampling of eroded material via pump samplers, bedload traps or natural sediment deposits risks being non-representative of the eroded material, due to selectivity in transport and deposition processes [*Owens et al.*, 2002]. Interrill erosion processes dominated sediment transport during the events, and are described in detail in Turnbull *et al*. [2010b]. Additional details of the experimental sites and summary metrics for the monitored rainfall events are provided in the Supporting Information (SI Figure 1; SI Table 1); for full description of the design and instrumentation of the plots, see *Turnbull et al.* [2010b, 2010a, 2011], *Puttock et al.* [2012, 2013, 2014] and *Brazier et al.* [2013].

2.3. Laboratory Analysis

Investigations characterising the chemistry of soil fractionated by particle size commonly deliberately disperse aggregates [e.g., *Quiroga et al.*, 1996; *Six et al.*, 2002; *von Lützow et al.*, 2007; *Marzaioli et al.*, 2010]. However, detailed investigations by *Chenu and Plante* [2006] have shown that solid mineral and organic matter are broken apart before aggregate structures are fully dispersed, challenging the concept of primary particles as a measurable unit. Depending on the nature of the precipitation event and soil characteristics, significant proportions of soil can be eroded in aggregate forms [*Alberts and Moldenhauer*, 1981; *Loch and Donnollan*, 1983; *Egashira and Nakai*, 1987; *Beuselinck et al.*, 2000; *Hu and Kuhn*, 2014]. In the present study, some eroded particles were stable in water and during dry sieving, but dispersed following acid treatment, apparently due to the removal of calcium carbonate; this finding suggests that further artificial disaggregation would be inappropriate when investigating particle-associated chemical transport in this calcareous environmental context. Therefore, fractionation was by effective particle size, in accordance with previous investigations into the erosion-induced redistribution of particulate-associated chemicals [*Egashira and Nakai*, 1987; *Slattery and Burt*, 1997; *Lister*, 2007; *Lister et al.*, 2007; *Nadeu et al.*, 2011].

Bulk samples of near-surface soil were fractionated by density using flotation-sedimentation density separation in deionised water and the > 1 g cm^{-3} fraction was dried at 60°C to a constant weight. Samples were then divided into eight effective particle size classes by dry-sieving at one ϕ (Wentworth phi) intervals (>4, 4-2, 2-1, 1-0.5, 0.5-0.25, 0.25-0.125, 0.125-0.0625, <0.0625 mm). Dry sieving was employed to minimise potential losses of soluble OC arising from wet sieving [*Beauchamp and Seech*, 1990; *Sainju et al.*, 2003, 2011; *Lister*, 2007]; such losses could be significant given the very low OC concentrations indicated by previous work [*Lister*, 2007; *Puttock*, 2013; *Puttock et al.*, 2012; *Brazier et al.*, 2013]. To an extent, dry sieving (all samples mechanically shaken consistently for 10 minutes) disaggregates loosely aggregated particles, and increasing shaking duration (up to 30 minutes) showed no further changes in gravimetrically-determined PSD, indicating that 10 minutes of shaking had disaggregated all loosely aggregated particles. This treatment helps to reduce problems with possible re-aggregation due to wetting and drying during sample preparation. The largest size threshold was considered appropriate because some particles >4 mm have been observed to erode during high-energy rainstorm events, and the minimum particle size threshold of <0.0625 mm is considered appropriate for undispersed particles [*Lister*, 2007; *Michaelides et al.*, 2012] and to parameterise numerical simulations given current limitations in the representation of detachment, transport and deposition of cohesive silt and clay particles [*Wainwright et al.*, 2008; *Turnbull et al.*, 2010c].

All eroded sediment was recovered from the stock tank, dried at 60°C to a constant weight and dry-sieved to determine PSD gravimetrically. The remixed sediment was subsampled with a riffle splitter before later being fractionated by effective particle size into five size classes (>2, 2-0.5, 0.5-0.25, 0.25-0.0625, <0.0625 mm). Relative to the eight size classes employed for the characterisation of near-surface soil, eroded sediment was fractionated at a coarser resolution to correspond with the PSD resolution recorded for sediment eroded during all monitored events [*Puttock*, 2013]. Each size fraction was subjected to flotation-sedimentation density separation in a 1 g cm^{-3} medium, and the >1 g cm^{-3} fraction was dried at 60°C.

To quantify OC in samples of soil and eroded sediment, inorganic carbon was removed via acid digestion. 5 g of each particle size fraction was digested in 75 ml of 2M HCL for seven days, filtered through a 0.45 µm filter, and triple rinsed with 100 ml of deionised water [*Turnbull et al.*, 2008b; *Puttock et al.*, 2012; *Puttock*, 2013; *Brazier et al.*, 2013]. To obtain representative samples, each particle size fraction larger than 0.125 mm was homogenised and all fractions larger than 0.25 mm were ground manually so as to pass through a 0.25 mm screen [*Sainju et al.*, 2003; *Lukasewycz and Burkhard*, 2005; *Wang et al.*, 2012, 2014b, 2015]. The elemental concentration of OC remaining was determined via dry combustion in an Elemental Analyser (Thermo Scientific, Flash 2000). Absolute instrument precision (defined as the standard deviation of standard reference materials) was ±0.22%; replicate analysis on 11.3% of the samples yielded a median relative difference in carbon concentration of just 6.1±1.9%, indicating aliquots were representative. In total, 592 unique samples were analysed.

2.4. Data Preparation and Statistical Analysis

Using size-sorted samples has been found to be more accurate than bulk samples for measuring total sediment-bound chemical pools when only small aliquots are analysed [*Michaelides et al.*, 2012]. Whole-soil OC concentrations were calculated by multiplying size-specific OC concentrations by the fractional mass of particles in each size class and summing values across sizes. Average OC concentrations [mass/mass, expressed as a %] and PSD for each surface cover (bare, grass, shrub) were weighted by fractional canopy cover (Table 1) to derive areally-weighted values for each site [after *Müller et al.*, 2007]. Near-surface (0-0.05 m) OC stocks [g m^{-2}] were calculated using the areally-weighted OC concentration for each site (expressed as a proportion), multiplied by areally-weighted bulk density [g m^{-3}] and sample depth [0.05 m].

OC event yields were determined by multiplying the observed particle size-specific OC concentration by mass eroded for each event. Although the near-surface soil samples were complete, 19/37 of the eroded sediment subsamples contained no coarse (>2 mm) particles; an omission arising from the low abundance of this size fraction in the original material, combined with limited subsample size. Because hillslope processes in these semi-arid ecosystems exhibit high degrees of inter-event variability [*Turnbull et al.*, 2010b, 2011, 2013; *Puttock et al.*, 2013; *Brazier et al.*, 2013], large ensembles of events are valuable to improve signal-to-noise ratios to support inferences regarding the mechanistic functioning of these ecosystems [as demonstrated by *Petrie et al.*, 2015]. To best use the available event ensemble, the 19 missing >2 mm OC concentrations were replaced with median >2 mm OC concentrations derived from each plot. This error introduced by this substitution is likely to be very small, because (i) particles of this size fraction comprised a small proportion (median 5%) of the overall PSD of eroded material, and (ii) variance in observed OC concentrations of this particle size fraction within each plot was not large (coefficient of variance ~30%). OC enrichment (ER_{OC}) can be expressed as the ratio of OC concentration in eroded soil (ES_{OC}) to that in the contributing soil (CS_{OC})

$$ER_{OC} = \frac{ES_{OC}}{CS_{OC}} \tag{1}$$

OC enrichment ratios were calculated for each particle size fraction and the total mass of eroded sediment for each event. To examine the extent to which particle size selectivity explains observed OC enrichment in eroded sediment, three OC event yields were calculated: (i) $\sum_{Obs}$ is the observed size-specific OC event yield, determined by multiplying the observed

OC concentration and mass of each particle size fraction eroded during each event, (ii) $\sum_{All}$ is the expected OC event yield, calculated using the average OC concentration of the contributing soil multiplied by the mass of eroded sediment, and (iii) $\sum_{PSD}$ is the expected OC event yield, calculated by summing the average OC concentration of the contributing soil for each particle size fraction by plot multiplied by the eroded mass of that fraction [*Palis et al.*, 1990b]. Assuming OC enrichment due to size selectivity *within* particle size fractions is minimal compared with OC enrichment due to size selectivity *between* particle size fractions, calculation of $\sum_{Obs}$, $\sum_{All}$ and $\sum_{PSD}$ enables calculation of the proportion of OC enrichment due to size selective transport (ER_{OC_PSD}), which can be expressed as

$$ER_{OC\_PSD} = \frac{\sum_{Obs} - \sum_{All}}{\sum_{PSD} - \sum_{All}} \tag{2}$$

Equation (2) is the ratio of observed enrichment to the enrichment predicted due to particle size selectivity. We also explored whether ER_{OC} was related to overall sediment concentration [e.g. *Wang et al.*, 2014a], were the total sediment concentration during each event (C_{event}) [g l^{-1}] was calculated as the total sediment yield (S_{event}) [g] normalised by the total runoff (Q_{event}) [l]

$$C_{event} = \frac{S_{event}}{Q_{event}} \tag{3}$$

Statistical analyses were conducted using R [*R Core Team*, 2015], and unless otherwise stated all errors are standard errors (SE). Results from the two grass-dominated and the two shrub-dominated sites were combined for heteroscedastic *t*-tests [see discussion in *Brazier et al.*, 2013].

3. Results

3.1. OC stocks in near-surface (0-0.05 m) soil

Four hundred aliquots were analysed to characterised OC concentrations in the near-surface soil. In addition to the expected peak in OC concentration in the finest (<0.0625 mm) fraction, there was a peak in some sand (1-0.5 mm and 2-1 mm) fractions; this bimodal distribution was consistent in all of the average values for each surface cover type at all sites. Across the grass-shrub ecotone, there was generally an overall decrease in the proportion of particles smaller than 0.125 mm and an increase in the proportion of particles larger than 0.25 mm (Figure 1~~Figure 1~~b). One hundred >2 mm aliquots were analysed, revealing OC concentrations ranging from 0.2% to 3.7% and <0.1% to 1.1% for the >4 mm and 4-2 mm fractions, respectively. The areally-weighted average OC concentration was very similar to the average OC concentrations of the fine (< 2 mm) fraction (Figure 2~~Figure 2~~a). These averages represent a wide range of concentrations, and are not an artefact caused by the lower detection limit of the elemental analyser.

The areally weighted, whole-soil, near-surface (0-0.05 m) OC stock is 275.8±24.0, 315.5±34.6, 390.5±60.8 and 327.7±36.3 g OC m^{-2}, in the grass-, grass-shrub-, shrub-grass- and shrub-dominated plots, respectively (Figure 2~~Figure 2~~b). Coarse (>2 mm) particles contribute 24% to 38% of these overall SOC stocks, mainly due to the abundance of these fractions (20% to 37% by weight) (Figure 1~~Figure 1~~c). The proportion of the total SOC stock associated with the coarsest (>4 mm) fraction increases across the grass-shrub ecotone, mainly due to changes in PSD (Figure 1~~Figure 1~~b and c). Despite its relatively low OC concentration, the 0.125-0.0625 mm fraction contributes substantially (18% to 22%) towards the whole-soil SOC stock, primarily due to the abundance of particles in this size-fraction (21% to 33% by weight) (Figure 1~~Figure 1~~b and c).

3.2. Erosion-induced OC event yields and enrichment dynamics

Observed OC event yields greatly exceeded those predicted using the average OC concentrations of the contributing surface soils, indicating substantial OC enrichment. The magnitude of the underprediction error is correlated with event yield magnitude, and the median underestimate was 65% (±4.9%). It is more appropriate to report mean event yield (± standard error) of OC rather than the total mass of eroded organic carbon for two reasons: (i) the convective rainfall which drives these erosion events is characteristically highly variable in both space and time [*Wainwright*, 2005; *Petrie et al.*, 2014]. Establishing these runoff plots across a vegetation ecotone in a natural ecosystem meant that the runoff plots could not be located immediately adjacent to one another, and while they were located within just a few km of each other the different plots therefore experienced different storm events over the monitoring periods [*Turnbull et al.*, 2010b]. (ii) Due to equipment limitations in these very harsh environments, it was not possible to measure the erosion-induced OC yields resulting from all erosion events. Critically however, in terms of total rainfall, total runoff, runoff coefficients and total sediment event yield, the 37 events presented herein are representative of all of the events observed over the four monsoon periods, albeit with some larger differences in the shrub-grass plot due to the small sample size analysed for OC yields (n=4) (Supporting Information – SI Figure 2). Mean OC event yield increased substantially across the grass-shrub ecotone, from 15.3, 22.2, 49.7 and 83.3 g from the grass, grass-shrub, shrub-grass and shrub dominated plots, respectively. The six-fold increase was caused by both (i) increasing erosion and (ii) increasing OC enrichment in the eroded sediment. A heteroscedastic t-test suggested that the difference in mean OC event yield between the two combined grass-dominated sites versus the two combined shrub-dominated sites was only statistically significant to the 6% level (t-2.034, p=0.059). OC event yields were very variable, both between events and between sites, with the standard error of the mean increasing across the grass-shrub ecotone from 7.8, 7.9, 24.5 and 36.7, for the grass, grass-shrub, shrub-grass and shrub sites respectively (Figure 3~~Figure 3~~a). The <0.25 mm particle size fractions contributed an average of 85.1% (±1.6%) of the total OC event yield over all events. Considering all sites together, event ER_{OC} values ranged from 1.0 to 10.2, and were greater than unity in 97% of the events, >2 in 68% of events, and >6 in 24% of events (Figure 3~~Figure 3~~b). Overall, ER_{OC} was statistically significantly >2 (Wilcoxon one-sample signed rank test; V=551, p<0.001). Stratifying by site reveals a substantial increase in mean OC enrichment across the grass-shrub ecotone, with mean ER_{OC} increasing from 2.74, 3.36, 4.89 and 5.16 for the grass, grass-shrub, shrub-grass and shrub dominated sites, respectively (Figure 3~~Figure 3~~b). Variation in ER_{OC} also increases across the grass-shrub transition, with SE increasing from 0.51, 0.73, 0.74 and 1.13 for the grass, grass-shrub, shrub-grass and shrub dominated sites, respectively. A heteroscedastic t-test indicated the difference between the two amalgamated grass-dominated and the two amalgamated shrub-dominated sites was statistically significant (t-2.126, p=0.044), with mean ER_{OC} of 3.04 and 5.09, respectively. OC enrichment was observed in all five particle size fractions during nearly all events, and across the grass-shrub ecotone, there was an increase in ER_{OC} in all particle size fractions smaller than 2 mm. In events showing overall OC enrichment (36/37), changes in PSD were found to explain a median average of 6% and up to 67% of observed OC enrichment. ER_{OC} was plotted against metrics of event intensity and magnitude: total rainfall, peak rainfall intensity, runoff coefficient, peak runoff, total runoff, total sediment event yield, total event sediment concentration (Figure 4); which did not indicate any strong relationships.

4. Discussion

4.1. PSD

Across the grass-shrub ecotone there is a decreasing proportion of <0.125 mm particles and an increasing proportion of >0.25 mm particles remaining in the near-surface soil. If it is assumed that changes in PSD observed in space across the grass-shrub ecotone represent change through time, this finding is consistent with the progressive degradation of the soil resource and development of stone pavement cover concomitant with vegetation change in this desert landscape [*Wainwright et al.*, 1995, 1999, 2000; *Turnbull et al.*, 2008a; *Michaelides et al.*, 2009; *Brazier et al.*, 2013; *Puttock et al.*, 2014].

4.2. Near-surface OC stocks

Both of the coarse (>4 mm and 4-2 mm) particle size fractions contained areally weighted mean OC concentrations similar to the fine (<2 mm) fraction (Figure 2~~Figure 2~~a), and accounted for 24% to 38% of the total near-surface SOC stocks (Figure 1~~Figure 1~~c). The proportion of the near-surface SOC stocks associated with the coarse particles cannot be simply extrapolated to deeper soil layers because erosion of fine particles by aeolian and fluvial processes can increase the relative abundance of coarse particle in the near-surface soil [*Larney et al.*, 1998; *Wainwright et al.*, 1999, 2000]. Critically, widely used standard protocols discard the >2 mm clasts, assuming they contain no SOC [*Robertson and Paul*, 2000; *Lal and Kimble*, 2001; *Ellert et al.*, 2001; *Bird et al.*, 2002; *Jackson et al.*, 2002; *Ewing et al.*, 2007; *Throop et al.*, 2012; *Sankey et al.*, 2012; *Frank et al.*, 2012; *Brazier et al.*, 2013; *Puttock et al.*, 2013, 2014]. Ignoring OC in the coarse fraction of these calcareous soils therefore risks substantial underestimation of SOC stocks in carbon inventories [sensu *Agnelli et al.*, 2002; *Corti et al.*, 2002].

Noteworthy concentrations of organic carbon in >2 mm clasts were also reported by Corti *et al*. [2002] and Agnelli *et al.* [2002] for a variety of environmental contexts, which they attributed to a combination of organic particles incorporated during the formation of sedimentary rocks and to subsequent infilling of porous rock fragments by soil solutions containing organic substances. These rock fragments contributed up to 4.5% of the total SOC in a forest soil, and were found to be chemically and biologically active in the soil, forming what they described as a continuum with the fine earth [*Agnelli et al.*, 2002].

In calcareous soils, the precipitation of calcium carbonate is known to stabilize soil aggregates [*Bryan*, 2000; *Nash and McLaren*, 2003; *Alonso-Zarza and Wright*, 2010], and in the present study substantial disaggregation was frequently observed in both soil and eroded sediment samples following the acid treatment, resulting in particle size reductions of up to five ϕ intervals in individual aggregates. Such stabilized aggregates are likely to include OC associated with fine particles, or fine particulate organic matter (POM) [*Duchaufour*, 1976; *Goudie*, 1996; *Baldock and Skjemstad*, 2000], particularly as the biochemical actions of roots and fungi facilitate calcium carbonate precipitation in arid soils [*Goudie*, 1996; *Alonso-Zarza and Wright*, 2010; *Gocke et al.*, 2011]. Therefore, it is argued here that the relatively substantial OC concentrations observed in coarse (>2 mm) particles are likely due to the stabilization of soil aggregates by precipitated calcium carbonate [*Duchaufour*, 1976; *Oyonarte et al.*, 1994; *Goudie*, 1996; *Baldock and Skjemstad*, 2000]. Calcium carbonate precipitation in calcareous dryland soils may contribute to the physical protection of OM from decomposition, both by forming thin coatings of pedogenic (secondary) carbonate on OM, and by stabilizing aggregates [*Duchaufour*, 1976; *Oyonarte et al.*, 1994; *Olk et al.*, 1995; *Baldock and Skjemstad*, 2000; *Clough and Skjemstad*, 2000; *Lopez-Sangil and Rovira*, 2013].

Consequently, it appears appropriate to recommend a re-evaluation of the ubiquitous assumption that the coarse (>2 mm) fraction of the soil is free of OC, particularly in environments with stabilised aggregates such as calcareous soils. While there is an extensive literature on many aspects of carbonate formation [*Breecker et al.*, 2009; *Alonso-Zarza and Wright*, 2010], and several studies mention the mechanisms by which precipitated calcium carbonate physically protects organic carbon [e.g., *Duchaufour*, 1976; *Oyonarte et al.*, 1994; *Olk et al.*, 1995; *Baldock and Skjemstad*, 2000; *Clough and Skjemstad*, 2000; *Lopez-Sangil and Rovira*, 2013], there appears to be something of a knowledge gap regarding the full implications of calcium carbonate precipitation for SOC dynamics in drylands. This mechanism may contribute to the characteristically high mean residence times of SOC in dryland ecosystems [*Frank et al.*, 2012], and radioisotope analysis could be utilised to determine whether organic carbon in the coarse (>2 mm) particle fraction is chemically and biologically active, as found by Agnelli *et al.* [2000, 2002] in a temperate forested environmental context.

4.3. Erosion-induced OC event yield and enrichment dynamics

This study describes the predominantly interrill erosion-induced efflux of OC from four large (300 m^2) runoff sites during 37 rainstorm-runoff events over a four year period. The analysis expands upon previous investigations into erosional carbon dynamics at this site [*Puttock et al.*, 2012, 2013, 2014; *Brazier et al.*, 2013] by quantifying the temporally-variable OC event yield through both wetter- and drier-than-average monsoon seasons [*Petrie et al.*, 2014], and represents the largest plot-scale characterisation of erosion-induced OC yields from any dryland ecosystem. This information is valuable because it represents total capture of sediment eroded from unperturbed sites during natural rainfall events for 31/37 erosion events, and substantial capture of sediment eroded during the other six events. Consequently, these data afford a more accurate representation of erosion-induced redistribution of OC in semi-arid natural landscapes than is possible using the predominantly laboratory-scale, reductionist experiments undertaken to date. The relatively long-scale monitoring is valuable in that it yields ensembles of natural erosion events, analyses of which helps to elucidate emergent properties of resource redistribution processes in these ecosystems.

The six-fold increase in average erosion-induced OC yields across the grass-shrub ecotone was driven predominantly by greater soil erosion (~3.5 fold increase) [for detailed discussion see *Jin et al.*, 2008, 2009; *Turnbull et al.*, 2010b], which is largely attributed to reduced vegetation cover and greater hydrological connectivity in the shrublands [*Turnbull et al.*, 2010b; *Puttock et al.*, 2013]. However, eroded sediments were also significantly enriched in OC relative to the contributing near-surface soil, and OC enrichment increased significantly across the grass-shrub ecotone, almost doubling from the grass-dominated plot to the shrub-dominated plot.

Because OC concentrations usually decrease rapidly with depth, ER_{OC} values are sensitive to the sampled depth of the contributing surface soils [*Cogle et al.*, 2002; *Li et al.*, 2007]. The 0-0.05 m depth considered herein is shallower than is often considered [e.g., *Quinton et al.*, 2006], which should increase the OC concentration of the contributing soil (CS_{OC}) relative to the OC concentration of the eroded soil (ES_{OC}), and therefore reduce ER_{OC} values. In contrast to this expectation, observed ER_{OC} were often far greater than the highest values commonly reported in the literature [e.g., ≤4.3 *Cogle et al.*, 2002; ≤5 *Lal*, 2003, 2005; ≤3 *Rhoton et al.*, 2006; ≤5.5 *Quinton et al.*, 2006; ≤2.2 *Truman et al.*, 2007; ≤2.2 *Jin et al.*, 2008; ≤3.9 *Wang et al.*, 2014a].

OC enrichment is commonly attributed to the selective detachment and transport of fine OC-rich particles [e.g., *Nelson et al.*, 1994; *Balesdent et al.*, 1998; *Guibert et al.*, 1999; *Rhoton et al.*, 2006; *Wang et al.*, 2013a]. Although the interrill erosion from our sites was strongly size-selective with preferential transport of fractions smaller than 0.25 mm [*Turnbull et al.*, 2010b; *Puttock*, 2013], differences in OC concentration between particle size fractions in the contributing soil were fairly small. Particle size selectivity was found to only explain an average of 6% of observed OC enrichment across the event ensemble; indicating changes in particle size selectivity do not significantly drive the significant, systematic change in ER_{OC} observed across the grass-shrub ecotone. Relative to black grama grasses, creosotebush shrubs produce more litter, which may also be more resistant to decay [*Liao et al.*, 2006a]. We hypothesize that these differences in the biotic processes continually contributing OC to the soil surface, which may not be incorporated evenly throughout the 0-0.05 m layer, causes an increased availability of OC in the uppermost surface soil of shrublands relative to grasslands and may therefore contribute to the observed increase in ER_{OC} across the grass-shrub ecotone.

Previous understanding arising from reductionist experimental work predicts that enrichment ratios should decrease over time towards unity, due to depletion of OC-rich fines in the source soil [e.g., *Polyakov and Lal*, 2004b; *Jin et al.*, 2009; *Hu et al.*, 2013]. However, there was no clear evidence of decreasing enrichment ratios over the four year study period, indicating that the previous finding may be an artefact of the experimental designs deployed in lab-based studies. The results presented herein suggest that OC enrichment can be an enduring phenomenon, at least at hillslope scales in semi-arid rangelands, and we believe that the preferential removal of OC may be sustained long-term by the dynamic replacement of OM via litter inputs via the soil surface [*Harden et al.*, 1999; *Li et al.*, 2007; *Berhe et al.*, 2008; *Doetterl et al.*, 2012]. This interpretation is consistent with monitoring of eight 875 m^2 runoff plots in an intensively managed temperate agroecosystem, which also found no decreasing trend in OC enrichment in the eroded sediments over a ten year monitoring period [*Quinton et al.*, 2006].

OC enrichment dynamics in eroded sediment may also be a function of rainfall intensity. Prior work suggests that OC enrichment will decrease during higher rainfall intensity, due to the increasing dominance of less-selective detachment and transport processes [*Ghadiri and Rose*, 1991b; *Truman et al.*, 2007; *Schiettecatte et al.*, 2008a; *Jin et al.*, 2009; *Wang et al.*, 2010, 2014a; *Kuhn et al.*, 2012], as discussed earlier (Section 1) and illustrated as Model 1 in Figure 5~~Figure 5~~a. However, in these semiarid ecosystems, changes in particle size selectivity are not so simply related to event magnitude, as larger rainstorm events produced smaller proportions of sand and a higher proportions of silt in eroded sediment [*Turnbull*, 2008; *Puttock*, 2013], and our results show that size-selective transport plays a minor role in OC enrichment. Instead, we hypothesize that the effect of rainfall intensity on OC enrichment can be modulated by spatial heterogeneity of soil characteristics, due to the possible concentration of fine and OC-rich particles in areas of higher topographic relief beneath vegetation, particularly shrubs [*Barth and Klemmedson*, 1978; *Schlesinger et al.*, 1990, 1996; *Kieft et al.*, 1998; *Wainwright et al.*, 2000; *Turnbull et al.*, 2010a; *Brazier et al.*, 2013; *Puttock et al.*, 2014; *Harman et al.*, 2014]. Previous work at these sites found that OC event yield was correlated with total event runoff and that the slope of this relationship steepened over the grass-shrub ecotone, indicating greater sensitivity of OC event yield to event runoff in the shrub-dominated plots [*Brazier et al.*, 2013]. Biogeochemical tracing of sediment eroded during a dryer-than-average period indicated that large proportions of the OC eroded from shrublands originated from bare interplant areas, where OC is older, legacy carbon from previously dominant grass vegetation [*Puttock et al.*, 2014], but that the proportion of shrub-derived OC associated with the eroded sediment increased during larger magnitude events [*Puttock*, 2013], a trend considered likely to continue during wetter periods.

Based on the above understanding, we propose refined conceptual models for OC enrichment as a function of rainfall intensity for grass-dominated (Model 2) and shrub-dominated (Model 3) hillslopes (

Figure 5~~Figure 5~~a). In grasslands, the (relatively) homogeneous distribution of OC results in low sensitivity of ER_{OC} to rainfall intensity. ER_{OC} is inversely related to rainfall intensity due to changes in the selectivity of dominant erosion processes; but always enriched, in contrast to Model 1. OC enrichment therefore occurs mainly due to the vertical gradient in OC concentrations within natural, non-homogenised soils (Model 2 (Grass) in

Figure 5~~Figure 5~~a). In shrublands, during low intensity rainfall erosion predominantly occurs in the bare interplant areas which have low soil OC concentrations relative to areas of microtopographic relief beneath vegetation [*Kieft et al.*, 1998; *Wainwright et al.*, 2000; *Brazier et al.*, 2013; *Harman et al.*, 2014] (Figure 5~~Figure 5~~b). Consequently eroded sediment may initially be depleted in OC ($ER_{OC} < 1$ in Figure 5~~Figure 5~~a). As rainfall intensity increases areas of topographic relief become inundated (Figure 5~~Figure 5~~b) with greater erosion of material from these OC-rich areas, enhancing OC enrichment in the eroded sediment (Model 3 in

Figure 5~~Figure 5~~a). The variable source areas caused by co-variation of topography and OC concentrations are hypothesised to produce a positive relationship between ER_{OC} and rainfall intensity, in contrast with understanding obtained from work in other, simpler, environmental contexts [cf. *Ghadiri and Rose*, 1991b; *Truman et al.*, 2007; *Schiettecatte et al.*, 2008a; *Wang et al.*, 2010, 2014a]. Our interpretation that differences in spatial distribution of OC concentrations across the grass-shrub ecotone [*Brazier et al.*, 2013] influence the OC concentration of eroded sediment and thus OC enrichment is consistent with the observation that inter-event variation in ER_{OC} increases across the grass-shrub ecotone. However, in the 37 storm ensemble presented herein, there were no consistent relationships between ER_{OC} and any individual metrics of rainfall event intensity or magnitude (total rainfall, peak 1-minute rainfall intensity, runoff coefficient, peak 1-minute runoff, total runoff, total sediment event yield, total event sediment concentration) (Figure 4). Therefore, we find no significant support for any of the three conceptual models described above, and we suggest that this finding reflects the low signal-to-noise ratios arising from the complex erosional dynamics of these natural ecosystems. Further elucidating controls on the OC enrichment dynamics of these complex natural hillslopes may require rainfall simulation experiments on natural hillslopes [e.g. *Parsons et al.*, 1997; *Truman et al.*, 2007] in order to increase control over variables such as antecedent conditions and rainfall intensities. This demonstrates the need for caution when extrapolating understanding from reductionist experiments to multifaceted real world environments [as acknowledged by *Wang et al.*, 2014a].

OC enrichment dynamics are a critical aspect of erosion-induced OC redistribution, and must therefore be represented in numerical models to accurately simulate erosion-induced OC fluxes [see *Polyakov and Lal*, 2004a; *Schiettecatte et al.*, 2008a]. While OC enrichment is typically attributed to size-selective detachment and transport, this process was negligible at our sites. Instead, we suggest that improvements in the predictive accuracy of deterministic models may require explicit consideration of topographic variation in OC concentration as influenced by surface cover (Figure 5~~Figure 5~~) and differences in transport dynamics associated with the lower density of OC-rich fractions.

While this study focuses on the erosion-induced redistribution of OC by overland flow processes, aeolian processes are acknowledged to be another key vector driving the redistribution of soil resources in dryland environments [*Larney et al.*, 1998; *Okin et al.*, 2004; *Li et al.*, 2007, 2008; *Ravi et al.*, 2007, 2010; *Field et al.*, 2010]. For example, monitoring aeolian erosion at the semi-arid Jornada Experimental Range in Southern New Mexico, USA, *Li et al.* [2007] found that up to 25% of the near-surface (0-0.05 m) soil OC stock was removed over three windy seasons, and that wind erosion-induced OC fluxes were inversely related with vegetation cover, due to accelerating erosion rates with reducing vegetation cover. They reported airborne sediments were enriched in OC by 3-6 times, relative to the contributing (0-0.05 m near-surface) soil, although further comparisons are hindered by the fact that their monitoring plots were somewhat disturbed by the vegetation removal treatments. Aeolian processes clearly play an important role in the redistribution of soil resources in semi-arid environments, and there is a need for co-located empirical studies to quantify concomitant fluxes arising from aeolian and fluvial processes [*Field et al.*, 2009;

Ravi et al., 2010]. Advancing mechanistic understanding of the interactions between aeolian and fluvial abiotic vectors will support their representation in numerical models used to elucidate emergent dynamics of complex ecosystems [see *Stewart et al.*, 2014].

Monitoring net ecosystem exchange of gaseous carbon has suggested shrub-dominated ecosystems take up significantly more carbon than grass-dominated ecosystems [*Petrie et al.*, 2015]. However, despite the higher rates of litter inputs to the soil surface from shrubs suggested by *Liao et al.* [2006b], we observe no difference in areal average, near-surface OC stocks across the grass-shrub ecotone, in agreement with previous studies [*Brazier et al.*, 2013; *Puttock et al.*, 2013]. The results presented here demonstrate that the erosion-induced OC yield is nearly six times higher from shrub-dominated sites relative to grass-dominated sites. Together, these findings indicate that shrub-dominated ecosystems appear to have a much quicker throughput of near-surface SOC relative to grass-dominated ecosystems. The substantial increase found in the erosion-induced yield of OC from shrub-dominated ecosystems compared with grass-dominated ecosystems implies that the higher net ecosystem exchange of gaseous carbon in shrublands relative to grasslands [*Petrie et al.*, 2015] does not invariably lead to increased sequestration of carbon in these terrestrial ecosystems [*Brazier et al.*, 2013]. Understanding the carbon sequestration potential of woody shrub encroachment requires comprehensive comparison of the carbon dynamics of grasslands versus shrublands [*Pacala et al.*, 2007; *Barger et al.*, 2011]. In addition to existing monitoring of gaseous fluxes [e.g., *Scott et al.*, 2009, 2016; *Petrie et al.*, 2015], this requires detailed understanding of erosion-induced carbon fluxes [*Li et al.*, 2007; *Brazier et al.*, 2013]. For example, *Wolkovich et al.* [2009] looked at carbon dynamics following grass encroachment into semi-arid shrubland, but acknowledged that their findings did not quantify potential changes in erosional fluxes arising from the changes in vegetation structure. The <1 g cm^{-3} fraction of eroded sediment, including most leaf litter, may also comprise a substantial proportion of the total OC efflux arising from runoff [*Bianchi*, 2011], and should be considered in future work monitoring lateral transfers of carbon in these ecosystems.

5. Conclusions

Coarse (>2 mm) particles can contain substantial amounts of OC, accounting for up to 38% of the total SOC stock in the semi-arid soils studied; this is likely to be due to the incorporation of organic carbon into macroaggregates stabilized by precipitated calcium carbonate into water stable forms. Standard soil analysis protocols assume that the >2 mm 'mineral' fraction contains no OC, which may be causing significant underestimation of SOC stocks.

OC enrichment can increase the erosion-induced redistribution of OC by up to an order of magnitude at hillslope-scales, and average enrichment increases significantly across the ecotone from grass-dominated to shrub-dominated communities. Predictions of OC enrichment dynamics based on reductionist experiments appeared to transfer poorly to complex, real-world environments, and OC enrichment appeared to be an enduring feature of uncultivated semiarid ecosystems. OC enrichment is often attributed to particle size selectivity, yet changes in PSD explained very little of the observed OC enrichment.

Across the transition from grass-dominated to shrub-dominated ecosystems there was a six-fold increase in the erosion-induced OC yields, due to both accelerated erosion and increased OC enrichment. Shrub-dominated ecosystems may have a quicker throughput of near-surface SOC relative to grass-dominated ecosystems, which suggests that higher net ecosystem exchange of gaseous carbon in shrublands relative to grasslands may not necessarily lead to increased sequestration of carbon in these ecosystems.

Acknowledgements

This research was conducted whilst A.M. Cunliffe was in receipt of a NERC Doctoral Training Grant (NE/K500902/1) and was supported by the NSF Long Term Ecological Research Program at the Sevilleta National Wildlife Refuge (DEB-1232294) and travel bursaries from the University of Sheffield, the Worshipful Company of Farmers, and the Royal Society Dudley Stamp Memorial Fund Award. Data presented herein is archived in the Sevilleta Data Repository (http://sev.lternet.edu), and with the corresponding author (AMC). We are grateful to John Buffington, Jon Pelletier and two anonymous reviews whose suggestions greatly improved upon earlier versions of this paper.

Table 1. Fractional canopy cover for all sites, derived from manual classification of near-ground aerial imagery [after *Turnbull et al.*, 2010a; *Puttock et al.*, 2013]. Photos by the author (July 2013).

Surface Cover	**Site**			
	Grass	**Grass-Shrub**	**Shrub-Grass**	**Shrub**
Bare	45.46%	57.00%	73.80%	79.35%
Grass	54.54%	38.60%	14.30%	0.00%
Shrub	0.00%	4.40%	11.90%	20.65%
Slope	4%	5%	7%	3%
Site Photo				

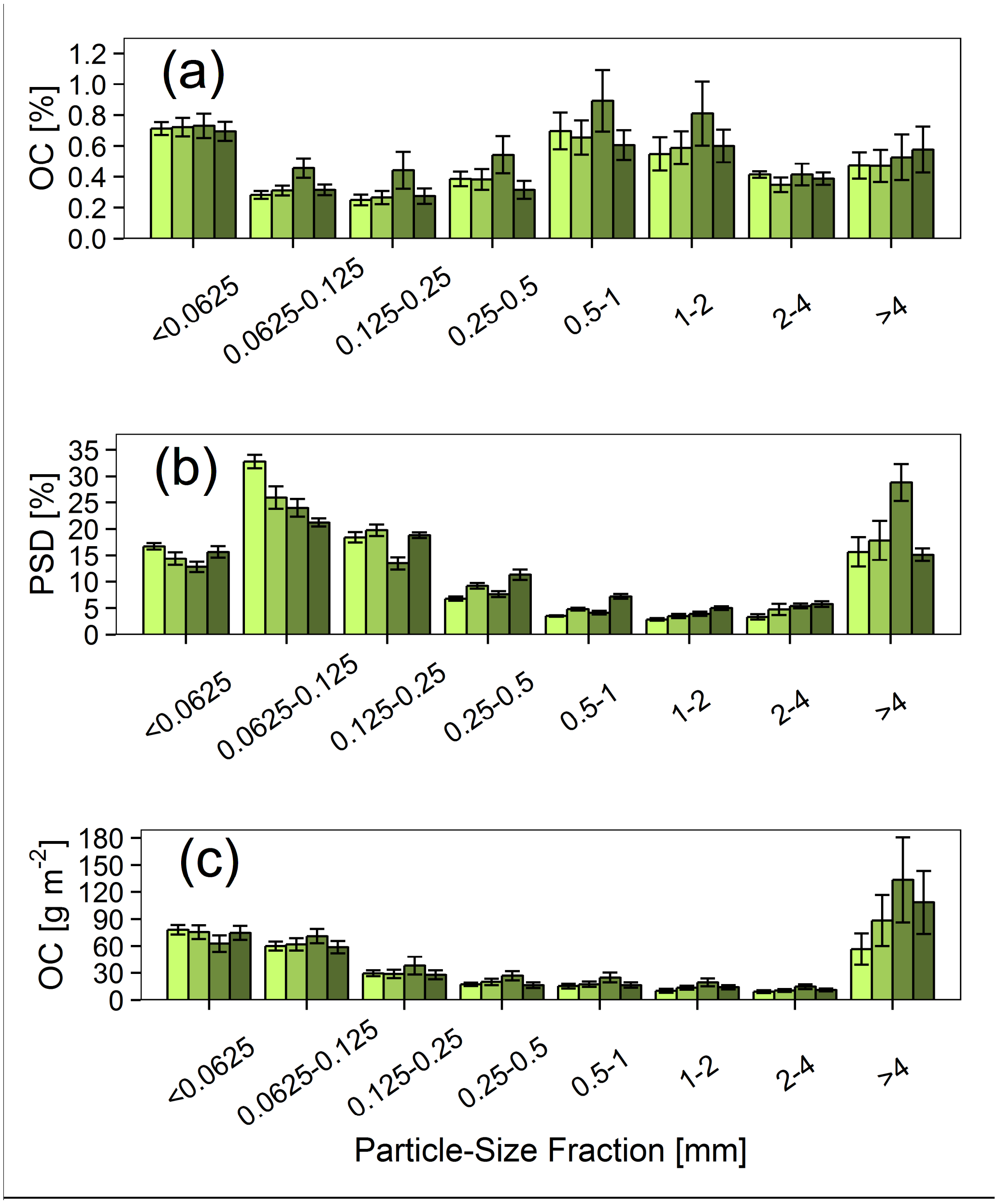


Figure 1. For each study plot across the grass-shrub ecotone: (a) areally-weighted organic carbon (OC) concentrations observed in each particle size fraction, (b) areally-weighted particle size distribution (PSD), and (c) areally-weighted OC concentration in each particle size fraction in near-surface (0-0.05 m) soil [g m^{-2}] (weighted by the fractional mass of each particle size fraction). Bar colours correspond to sites across the grass shrub ecotone (as shown in Table 1). Values are means ± standard error.

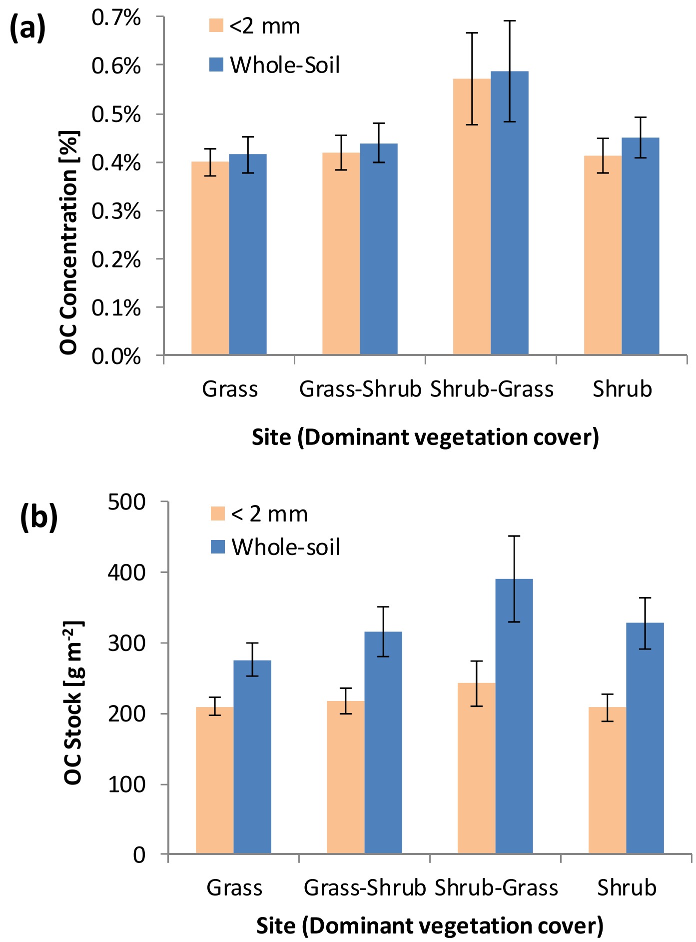


Figure 2. (a) Areally-weighted near-surface (0-0.05 m) average organic carbon (OC) concentrations in the <2 mm fraction and whole soil. (b) Areally-weighted near-surface soil organic carbon (OC) stocks for each site, calculated for the <2 mm fraction, and with the whole-soil OC concentration (including the >2 mm fractions). Values are means ± standard error.

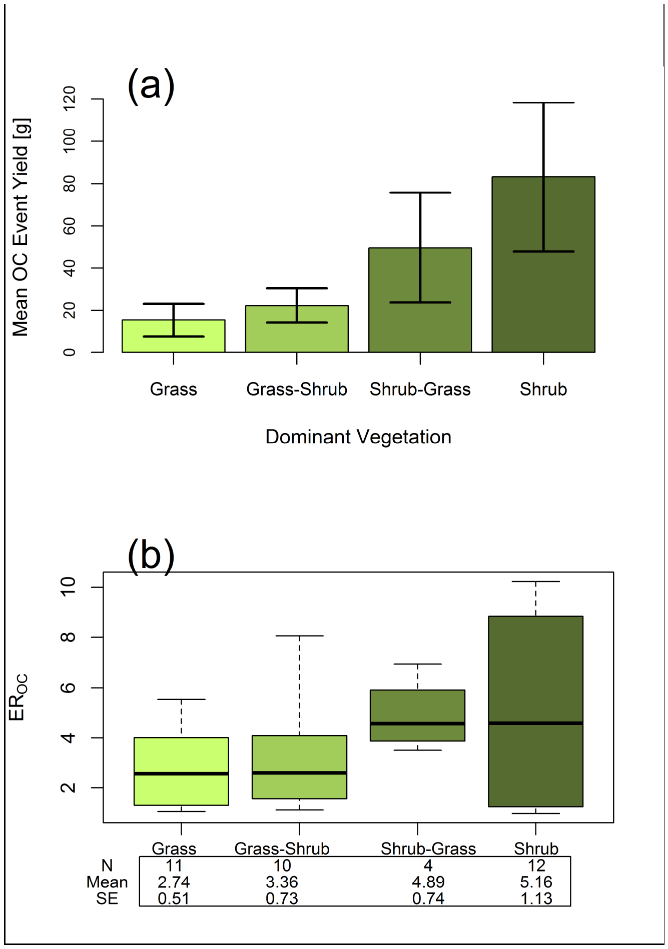


Figure 3. (a) Mean organic carbon (OC) event yield (± standard error) and (b) OC enrichment ratios and summary statistics, stratified by site. Where N is number of rainfall events, and SE is standard error. Bar colours correspond to sites across the grass shrub ecotone (as shown in Table 1). In the boxplots, from top to bottom, horizontal bars represent the maximum, upper quartile, median, lower quartile, and minimum values.

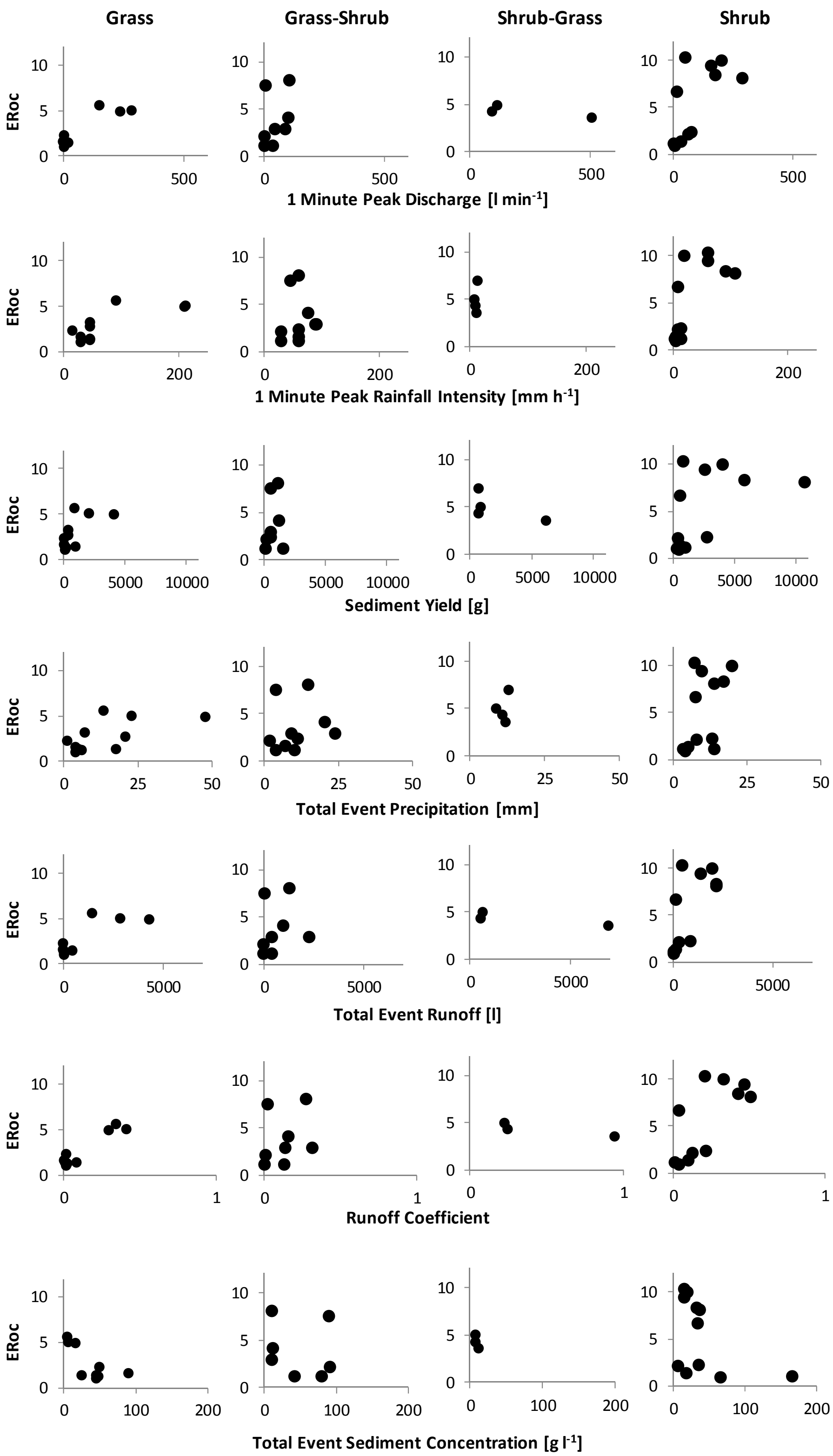


Figure 4. Relationships between the organic carbon enrichment ratio (ER_{OC}) and metrics of event intensity and magnitude: 1 minute peak discharge, 1 minute peak rainfall intensity, sediment yield, total event precipitation, total runoff, runoff coefficient, and total event sediment concentration ($C_{event} = S_{event} / Q_{event}$).

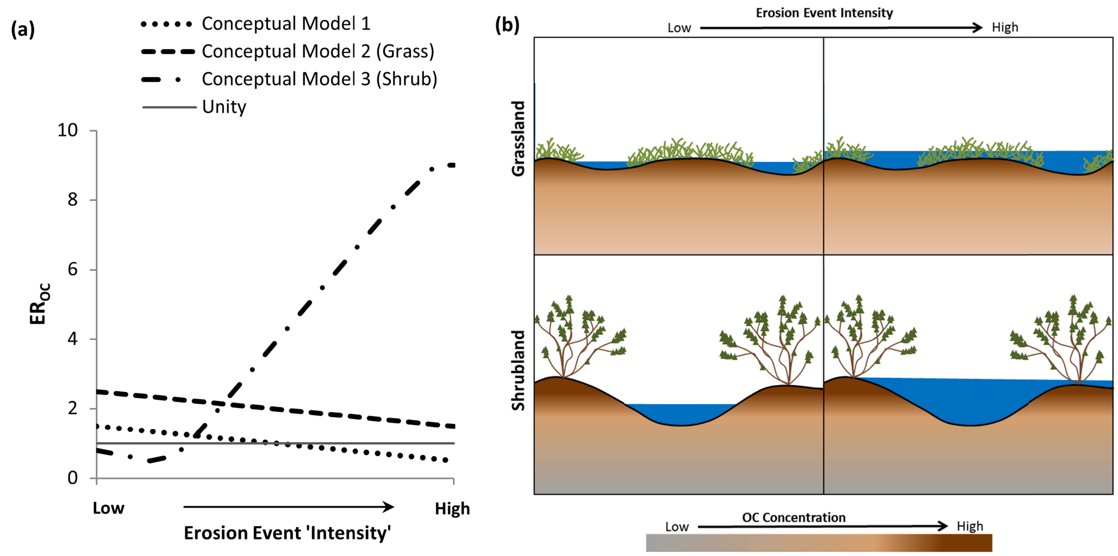


Figure 5. (a) Conceptual models of the relationship between event intensity and the enrichment ratio of organic carbon (ER_{OC}). Conceptual Model 1 reflects understanding from previous laboratory experiments documenting changes in sediment source areas (interrill vs. rill) and associated degree of size-selective transport of OC-rich fines with increasing rainfall intensity [discussed in *Schiettecatte et al.*, 2008a]. Conceptual Model 2 (Grass) is the authors' expectation for these grass-dominated ecosystems based on understanding of the (relatively) homogeneous distribution of OC and topography. Conceptual Model 3 (Shrub) is the authors' expectation for these shrub-dominated ecosystems based on understanding of the heterogeneous, co-varying distribution of OC and topography. (b) Schematic representation of how differences in the microtopography and spatial distribution of OC between grass-dominated ecosystems (upper) and shrub-dominated ecosystems (lower) influence the availability to erosion of soil OC arising from different depths of overland flow – higher OC concentrations are indicated by darker brown shading.

References

Adeel, Z., U. Safriel, D. Niemeijer, and R. White (2005), *Ecosystems and Human Wellbeing: Desertification Synthesis*, Millennium Ecosystem Assessment, World Resources Institute, Washington, D.C.

Agnelli, A., L. Celi, A. Degl'Innocenti, G. Corti, and F. C. Ugolini (2000), Chemical and spectroscopic characterization of the humic substances from sandstone-derived rock fragments, *Soil Sci.*, *165*(4), 314–327.

Agnelli, A., S. E. Trumbore, G. Corti, and F. C. Ugolini (2002), The dynamics of organic matter in rock fragments in soil investigated by 14C dating and measurements of 13C, *Eur. J. Soil Sci.*, *53*(1), 147–159, doi:10.1046/j.1365-2389.2002.00432.x.

Ahlström, A. et al. (2015), The dominant role of semi-arid ecosystems in the trend and variability of the land CO2 sink, *Science*, *348*(6237), 895–899, doi:10.1126/science.aaa1668.

Alberts, E. E., and W. C. Moldenhauer (1981), Nitrogen and phosphorus transported by eroded soil aggregates, *Soil Sci. Soc. Am. J.*, *45*(2), 391, doi:10.2136/sssaj1981.03615995004500020032x.

Alonso-Zarza, A. M., and V. P. Wright (2010), Chapter 5 Calcretes, in *Developments in Sedimentology*, vol. 61, edited by A. M. A.-Z. and L. H. Tanner, pp. 225–267, Elsevier.

Baldock, J. A., and J. O. Skjemstad (2000), Role of the soil matrix and minerals in protecting natural organic materials against biological attack, *Org. Geochem.*, *31*(7–8), 697–710, doi:10.1016/S0146-6380(00)00049-8.

Balesdent, J., E. Besnard, D. Arrouays, and C. Chenu (1998), The dynamics of carbon in particle-size fractions of soil in a forest-cultivation sequence, *Plant Soil*, *201*, 49–57, doi:10.1023/A:1004337314970.

Barger, N. N., S. R. Archer, J. L. Campbell, C. Huang, J. A. Morton, and A. K. Knapp (2011), Woody plant proliferation in North American drylands: a synthesis of impacts on ecosystem carbon balance, *J. Geophys. Res.*, *116*, G00K07, doi:10.1029/2010JG00150.

Barnes, P. W., H. L. Throop, D. B. Hewins, M. L. Abbene, and S. R. Archer (2012), Soil coverage reduces photodegradation and promotes the development of soil-microbial films on dryland leaf litter, *Ecosystems*, *15*(2), 311–321, doi:10.1007/s10021-011-9511-1.

Barth, R. C., and J. O. Klemmedson (1978), Shrub-induced spatial patterns of dry matter, nitrogen, and organic carbon, *Soil Sci. Soc. Am. J.*, *42*, 804–809.

Beauchamp, E. G., and A. G. Seech (1990), Denitrification with different sizes of soil aggregates obtained from dry-sieving and from sieving with water, *Biol. Fertil. Soils*, *10*(3), 188–193, doi:10.1007/BF00336134.

Beniston, J. W., M. J. Shipitalo, R. Lal, E. A. Dayton, D. W. Hopkins, F. Jones, A. Joynes, and J. a. J. Dungait (2015), Carbon and macronutrient losses during accelerated erosion under different tillage and residue management, *Eur. J. Soil Sci.*, *66*(1), 218–225, doi:10.1111/ejss.12205.

Berhe, A. A., J. W. Harden, M. S. Torn, and J. Harte (2008), Linking soil organic matter dynamics and erosion-induced terrestrial carbon sequestration at different landform positions, *J. Geophys. Res. Biogeosciences*, *113*, G04039, doi:10.1029/2008jg000751.

Bestelmeyer, B. T., G. S. Okin, M. C. Duniway, S. R. Archer, N. F. Sayre, J. C. Williamson, and J. E. Herrick (2015), Desertification, land use, and the transformation of global drylands, *Front. Ecol. Environ.*, *13*(1), 28–36, doi:10.1890/140162.

Beuselinck, L., A. Steegen, G. Govers, J. Nachtergaele, I. Takken, and J. Poesen (2000), Characteristics of sediment deposits formed by intense rainfall events in small catchments in the Belgian Loam Belt, *Geomorphology*, *32*, 69–82, doi:10.1016/S0169-555X(99)00068-9.

Bianchi, T. S. (2011), The role of terrestrially derived organic carbon in the coastal ocean: A changing paradigm and the priming effect, *Proc. Natl. Acad. Sci.*, *108*(49), 19473–19481, doi:10.1073/pnas.1017982108.

Bird, S. B., J. E. Herrick, M. M. Wander, and S. F. Wright (2002), Spatial heterogeneity of aggregate stability and soil carbon in semi-arid rangeland, *Environ. Pollut.*, *116*, 445–455.

Bouwman, A. F. (1989), Modelling soil organic matter decomposition and rainfall erosion in two tropical soils after forest clearing for permanent agriculture, *Land Degrad. Dev.*, *1*, 125–140.

Brazier, R. E., L. Turnbull, R. Bol, and J. Wainwright (2013), Carbon loss by water erosion in drylands: implications from a study of vegetation change in the southwest USA, *Hydrol. Process.*, doi:10.1002/hyp.9741.

Breecker, D. O., Z. D. Sharp, and L. D. McFadden (2009), Seasonal bias in the formation and stable isotopic composition of pedogenic carbonate in modern soils from central New Mexico, USA, *Geol. Soc. Am. Bull.*, *121*, 630–640.

Bryan, R. B. (2000), Soil erodibility and processes of water erosion on hillslope, *Geomorphology*, *32*(3–4), 385–415, doi:10.1016/S0169-555X(99)00105-1.

Chartier, M. P., C. M. Rostagno, and L. S. Videla (2013), Selective erosion of clay, organic carbon and total nitrogen in grazed semiarid rangelands of northeastern Patagonia, Argentina, *J. Arid Environ.*, *88*, 43–49, doi:10.1016/j.jaridenv.2012.08.011.

Chenu, C., and A. F. Plante (2006), Clay-sized organo-mineral complexes in a cultivation chronosequence: revisiting the concept of the "primary organo-mineral complex," *Eur. J. Soil Sci.*, *57*(4), 596–607, doi:10.1111/j.1365-2389.2006.00834.x.

Clough, A., and J. O. Skjemstad (2000), Physical and chemical protection of soil organic carbon in three agricultural soils with different contents of calcium carbonate, *Soil Res.*, *38*(5), 1005–1016.

Cogle, A. l., K. p. c. Rao, D. f. Yule, G. . Smith, P. j. George, S. t. Srinivasan, and L. Jangawad (2002), Soil management for Alfisols in the semiarid tropics: erosion, enrichment ratios and runoff, *Soil Use Manag.*, *18*(1), 10–17, doi:10.1111/j.1475-2743.2002.tb00044.x.

Conant, R. T., J. M. Klopatek, R. C. Malin, and C. C. Klopatek (1998), Carbon pools and fluxes along an environmental gradient in northern Arizona, *Biogeochemistry*, *43*, 43–61.

Corti, G., F. C. Ugolini, A. Agnelli, G. Certini, R. Cuniglio, F. Berna, and M. J. Fernández Sanjurjo (2002), The soil skeleton, a forgotten pool of carbon and nitrogen in soil, *Eur. J. Soil Sci.*, *53*(2), 283–298, doi:10.1046/j.1365-2389.2002.00442.x.

Doetterl, S., J. Six, B. Van Wesemael, and K. Van Oost (2012), Carbon cycling in eroding landscapes: geomorphic controls on soil organic C pool composition and C stabilization, *Glob. Change Biol.*, *18*, 2218–2232, doi:10.1111/j.1365-2486.2012.02680.x.

Duchaufour, P. (1976), Dynamics of organic matter in soils of temperate regions: Its action on pedogenesis, *Geoderma*, *15*(1), 31–40, doi:10.1016/0016-7061(76)90068-9.

Egashira, K., and S. Nakai (1987), Size distribution and wet density of sediment eroded under simulated rainfall, *Soil Sci. Plant Nutr.*, *33*(3), 347–354, doi:10.1080/00380768.1987.10557580.

Eldridge, D. J., M. A. Bowker, F. T. Maestre, E. Roger, J. F. Reynolds, and W. G. Whitford (2011), Impacts of shrub encroachment on ecosystem structure and functioning: towards a global synthesis, *Ecol. Lett.*, *14*, 709–722, doi:10.1111/j.1461-0248.2011.01630.x.

Ellert, B. H., H. H. Janzen, and B. G. McConkey (2001), Measuring and comparing soil carbon storage, in *Assessment Methods for Soil Carbon*, edited by R. Lal, J. M. Kimble, R. F. Follett, and B. A. Steward, pp. 131–146.

Ewing, S. A., R. J. Southard, J. L. Macalady, A. S. Hartshorn, and M. J. Johnson (2007), Soil microbial fingerprints, carbon, and nitrogen in a Mojave Desert creosote-bush ecosystem, *Soil Sci. Soc. Am. J.*, *71*, 469–475.

Field, J. P., D. D. Breshears, and J. J. Whicker (2009), Toward a more holistic perspective of soil erosion: Why aeolian research needs to explicitly consider fluvial processes and interactions, *Aeolian Res.*, *1*(1–2), 9–17, doi:10.1016/j.aeolia.2009.04.002.

Field, J. P., J. Belnap, D. D. Breshears, J. Neff, G. S. Okin, J. J. Whicker, T. H. Painter, S. Ravi, M. C. Reheis, and R. Reynolds (2010), The ecology of dust, *Front. Ecol. Environ.*, *8*, 423–430, doi:10.1890/090050.

Fierer, N. G., and E. J. Gabet (2002), Carbon and nitrogen losses by surface runoff following changes in vegetation, *J. Environ. Qual.*, *31*, 1207–1213, doi:10.2134/jeq2002.1207.

Finch, D. M. (2012), *Climate Change in Grasslands, Shrublands and Deserts of the Interior American West: A Review and Needs Assessment*, United States Department of Agriculture, Fort Collins.

Foereid, B., M. J. Rivero, O. Primo, and I. Ortiz (2011), Modelling photodegradation in the global carbon cycle, *Soil Biol. Biochem.*, *43*(6), 1383–1386, doi:10.1016/j.soilbio.2011.03.004.

Frank, D. A., A. W. Pontes, and K. J. McFarlane (2012), Controls on soil organic carbon stocks and turnover among North American ecosystems, *Ecosystems*, *15*, 604–615, doi:10.1007/s10021-012-9534-2.

Ghadiri, H., and C. W. Rose (1991a), Sorbed chemical transport in overland flow: I. A nutrient and pesticide enrichment mechanism, *J. Environ. Qual.*, *20*, 628–633.

Ghadiri, H., and C. W. Rose (1991b), Sorbed chemical transport in overland flow: II. enrichment ratio variation with erosion processes, *J. Environ. Qual.*, *20*, 634–641, doi:10.2134/jeq1991.00472425002000030021x.

Glenn, E., M. Stafford Smith, and V. Squires (1998), On our failure to control desertification: implications for global change issues, and a research agenda for the future, *Environ. Sci. Policy*, *1*(2), 71–78, doi:10.1016/S1462-9011(98)00007-0.

Gocke, M., K. Pustovoytov, and Y. Kuzyakov (2011), Carbonate recrystallization in root-free soil and rhizosphere of Triticum aestivum and Lolium perenne estimated by 14C labeling, *Biogeochemistry*, *103*(1-3), 209–222, doi:10.1007/s10533-010-9456-z.

Goodale, C. L., and E. A. Davidson (2002), Uncertain sinks in the shrubs, *Nature*, *418*, 593.

Goudie, A. S. (1996), Organic agency in calcrete development, *J. Arid Environ.*, *32*(2), 103–110, doi:10.1006/jare.1996.0010.

Guibert, H., P. Fallavier, and J.-J. Roméro (1999), Carbon content in soil particle size and consequence on cation exchange capacity of alfisols, *Commun. Soil Sci. Plant Anal.*, *30*, 2521–2537, doi:10.1080/00103629909370392.

Harden, J. W., J. M. Sharpe, W. J. Parton, D. S. Ojima, T. L. Fries, T. G. Huntington, and S. M. Dabney (1999), Dynamic replacement and loss of soil carbon on eroding cropland, *Glob. Biogeochem. Cycles*, *13*, 885–901, doi:10.1029/1999gb900061.

Harman, C. J., K. A. Lohse, P. A. Troch, and M. Sivapalan (2014), Spatial patterns of vegetation, soils and microtopography from terrestrial laser scanning on two semi-arid hillslopes of contrasting lithology, *J. Geophys. Res. Biogeosciences*, *119*(2), 163–180, doi:10.1002/2013JG002507.

Hu, Y., and N. J. Kuhn (2014), Aggregates reduce transport distance of soil organic carbon: are our balances correct?, *Biogeosciences Discuss*, *11*(6), 8829–8859, doi:10.5194/bgd-11-8829-2014.

Hu, Y., W. Fister, and N. J. Kuhn (2013), Temporal variation of SOC enrichment from interrill erosion over prolonged rainfall simulations, *Agriculture*, *3*(4), 726–740, doi:10.3390/agriculture3040726.

Jacinthe, P.-A., and R. Lal (2001), A mass balance approach to assess carbon dioxide evolution during erosional events, *Land Degrad. Dev.*, *12*(4), 329–339, doi:10.1002/ldr.454.

Jacinthe, P.-A., R. Lal, and J. M. Kimble (2001), Assessing water erosion impacts on soil carbon pools and fluxes, in *Assessment Methods for Soil Carbon*, edited by R. Lal, J. M. Kimble, R. F. Follett, and B. A. Stewart, pp. 427–449, Lewis Publishers, Boca Raton, FL, USA.

Jacinthe, P.-A., R. Lal, L. B. Owens, and D. L. Hothem (2004), Transport of labile carbon in runoff as affected by land use and rainfall characteristics, *Soil Tillage Res.*, *77*, 111–123, doi:10.1016/j.still.2003.11.004.

Jackson, R. B., J. L. Banner, E. G. Jobbágy, W. T. Pockman, and D. H. Wall (2002), Ecosystem carbon loss with woody plant invasion of grasslands, *Nature*, *418*, 623–626, doi:10.1038/nature00910.

Jin, K. et al. (2008), Redistribution and loss of soil organic carbon by overland flow under various soil management practices on the Chinese Loess Plateau, *Soil Use Manag.*, *24*(2), 181–191, doi:10.1111/j.1475-2743.2008.00151.x.

Jin, K. et al. (2009), Residue cover and rainfall intensity effects on runoff soil organic carbon losses, *Catena*, *78*(1), 81–86, doi:10.1016/j.catena.2009.03.001.

Kieft, T. L., C. S. White, S. R. Loftin, R. Aguilar, J. A. Craig, and D. A. Skaar (1998), Temporal dynamics in soil carbon and nitrogen resources at a grassland-shrubland ecotone, *Ecology*, *79*, 671–683.

Kuhn, N. J. (2007), Erodibility of soil and organic matter: independence of organic matter resistance to interrill erosion, *Earth Surf. Process. Landf.*, *32*, 794–802, doi:10.1002/esp.1486.

Kuhn, N. J., and E. K. Armstrong (2012), Erosion of organic matter from sandy soils: Solving the mass balance, *Catena*, *98*, 87–95, doi:10.1016/j.catena.2012.05.014.

Kuhn, N. J., E. K. Armstrong, A. C. Ling, K. L. Connolly, and G. Heckrath (2012), Interrill erosion of carbon and phosphorus from conventionally and organically farmed Devon silt soils, *Catena*, *91*, 94–103, doi:10.1016/j.catena.2010.10.002.

Kuzyakov, Y. (2010), Priming effects: Interactions between living and dead organic matter, *Soil Biol. Biochem.*, *42*(9), 1363–1371, doi:10.1016/j.soilbio.2010.04.003.

Lacoste, M., V. Viaud, D. Michot, and C. Walter (2015), Landscape-scale modelling of erosion processes and soil carbon dynamics under land-use and climate change in agroecosystems, *Eur. J. Soil Sci.*, n/a–n/a, doi:10.1111/ejss.12267.

Lal, R. (1976), *Soil erosion problems on an alfisol in western Nigeria and their control*, International Institute of Tropical Agriculture, Ibadan, Nigeria.

Lal, R. (2001), Potential of desertification control to sequester carbon and mitigate the greenhouse effect, *Clim. Change*, *51*, 35–72.

Lal, R. (2003), Soil erosion and the global carbon budget, *Environ. Int.*, *29*, 437–450, doi:10.1016/S0160-4120(02)00192-7.

Lal, R. (2005), Soil erosion and carbon dynamics, *Soil Tillage Res.*, *81*, 137–142, doi:10.1016/j.still.2004.09.002.

Lal, R., and J. M. Kimble (2001), Importance of soil bulk density and methods of its importance, in *Assessment Methods for Soil Carbon*, edited by R. Lal, J. M. Kimble, R. F. Follett, and B. A. Stewart, pp. 31–44.

Lal, R., and D. Pimentel (2008), Soil erosion: A carbon sink or source?, *Science*, *319*, 1040–1042, doi:10.1126/science.319.5866.1040.

Lal, R., R. F. Follett, and J. M. Kimble (2001), Research and development priorities, in *The Potential of US Grazing Lands to Sequester Carbon and Mitigate the Greenhouse Effect*, edited by R. F. Follett, J. M. Kimble, and R. Lal, pp. 231–238, Lewis Publishers, Boca Raton, FL, USA.

Lal, R., M. Griffin, J. Apt, L. Lave, and M. G. Morgan (2004), Managing soil carbon, *Science*, *304*, 393–393.

Larney, F. J., M. S. Bullock, H. H. Janzen, B. H. Ellert, and E. C. S. Olson (1998), Wind erosion effects on nutrient redistribution and soil productivity, *J. Soil Water Conserv.*, *53*(2), 133–140.

Lee, J. J., D. L. Phillips, and R. F. Dodson (1996), Sensitivity of the US corn belt to climate change and elevated CO2: II. Soil erosion and organic carbon, *Agric. Syst.*, *52*, 503–521, doi:10.1016/S0308-521X(96)00015-7.

Liao, J. D., T. W. Boutton, and J. D. Jastrow (2006a), Organic matter turnover in soil physical fractions following woody plant invasion of grassland: Evidence from natural 13C and 15N, *Soil Biol. Biochem.*, *38*, 3197–3210, doi:10.1016/j.soilbio.2006.04.004.

Liao, J. D., T. W. Boutton, and J. D. Jastrow (2006b), Storage and dynamics of carbon and nitrogen in soil physical fractions following woody plant invasion of grassland, *Soil Biol. Biochem.*, *38*, 3184–3196.

Li, J., G. S. Okin, L. J. Alvarez, and H. E. Epstein (2007), Quantitative effects of vegetation cover on wind erosion and soil nutrient loss in a desert grassland of southern New Mexico, USA, *Biogeochemistry*, *85*, 317–332, doi:10.1007/s10533-007-9142-y.

Li, J., G. S. Okin, L. J. Alvarez, and H. E. Epstein (2008), Effects of wind erosion on the spatial heterogeneity of soil nutrients in two desert grassland communities, *Biogeochemistry*, *88*, 73–88.

Lister, D. (2007), Small-Scale Erosion-Driven Nutrient Dynamics in Different Vegetation Communities in Jornada, New Mexico: Implications for Land Degradation, University of Bristol.

Lister, D., K. Michaelides, J. L. Wadham, J. Wainwright, and A. J. Parsons (2007), Erosion-driven nutrient dynamics in different vegetation communities in Jornada, New Mexico: implications for land degradation, in *Geophysical Research Abstracts*.

Loch, R., and T. Donnollan (1983), Field rainfall simulator studies on two clay soils of the Darling Downs, Queensland. II. Aggregate Breakdpwn, sediment properties and soil erodibility, *Soil Res.*, *21*(1), 47–58.

Lopez-Sangil, L., and P. Rovira (2013), Sequential chemical extractions of the mineral-associated soil organic matter: An integrated approach for the fractionation of organo-mineral complexes, *Soil Biol. Biochem.*, *62*, 57–67, doi:10.1016/j.soilbio.2013.03.004.

Lukasewycz, M. T., and L. P. Burkhard (2005), Complete elimination of carbonates: A critical step in the accurate measurement of organic and black carbon in sediments, *Environ. Toxicol. Chem.*, *24*(9), 2218–2221, doi:10.1897/04-653R.1.

von Lützow, M., I. Kögel-Knabner, K. Ekschmitt, H. Flessa, G. Guggenberger, E. Matzner, and B. Marschner (2007), SOM fractionation methods: Relevance to functional pools and to stabilization mechanisms, *Soil Biol. Biochem.*, *39*, 2183–2207, doi:10.1016/j.soilbio.2007.03.007.

Maestre, F. T., J. Reynolds, E. Huber-Sannwald, J. E. Herrick, and M. Stafford-Smith (2006), Understanding global desertification: biophysical and socioeconomic dimensions of hydrology, in *Dryland Ecohydrology*, edited by P. D'Odorico and A. Porporato, pp. 315–332, Springer.

Malam Issa, O., Y. L. Bissonnais, O. Planchon, D. T. Favis-Mortlock, N. Silvera, and J. Wainwright (2006), Soil detachment and transport on field- and laboratory- scale interrill areas: erosion processes and the size-selectivity of eroded sediment, *Earth Surf. Process. Landf.*, *31*, 929–939, doi:10.1002/esp.1303.

Marzaioli, F., C. Lubritto, I. D. Galdo, A. D'Onofrio, M. F. Cotrufo, and F. Terrasi (2010), Comparison of different soil organic matter fractionation methodologies: Evidences from ultrasensitive 14C measurements, *Nucl. Instrum. Methods Phys. Res. Sect. B Beam Interact. Mater. At.*, *268*, 1062–1066, doi:10.1016/j.nimb.2009.10.098.

Mayeux, H. (2001), Foreword, in *The Potential of U.S. Grazing Lands to Sequester Carbon and Mitigate the Greenhouse Effect*, edited by R. F. Follett, J. M. Kimble, and R. Lal, pp. XXV–XXIX, Lewis Publishers, Boca Raton, FL, USA.

Meixner, T., P. D. Brooks, J. Hogan, C. Soto, and S. Simpson (2012), Carbon and nitrogen export from semiarid uplands to perennial rivers: connections and missing links, San Pedro River, Arizona, USA, *Geogr. Compass*, *6*, 546–559.

Michaelides, K., D. Lister, J. Wainwright, and A. J. Parsons (2009), Vegetation controls on small-scale runoff and erosion dynamics in a degrading dryland environment, *Hydrol. Process.*, *23*(11), 1617–1630, doi:10.1002/hyp.7293.

Michaelides, K., D. Lister, J. Wainwright, and A. J. Parsons (2012), Linking runoff and erosion dynamics to nutrient fluxes in a degrading dryland landscape, *J. Geophys. Res. Biogeosciences*, *117*(G4), n/a–n/a, doi:10.1029/2012JG002071.

Mitchell, H. F., P. G. Lakshminarayan, T. Otake, and B. A. Babcock (1998), The impact of soil conservation policies on carbon sequestration in agricultural soils of the central United States, in *Management of Carbon Sequestration in Soil*, edited by R. Lal, J. M. Kimble, R. F. Follett, and B. A. Stewart, CRC Press, Boca Raton, FL, USA.

Mora, J. L., J. A. Guerra, C. M. Armas, A. Rodríguez-Rodríguez, C. D. Arbelo, and J. S. Notario (2007), Mineralization rate of eroded organic C in Andosols of the Canary Islands, *Sci. Total Environ.*, *378*, 143–146, doi:10.1016/j.scitotenv.2007.01.040.

Müller, E. N., J. Wainwright, and A. J. Parsons (2007), Impact of connectivity on the modeling of overland flow within semiarid shrubland environments, *Water Resour. Res.*, *43*, doi:10.1029/2006WR005006.

Nadeu, E., J. de Vente, M. Martínez-Mena, and C. Boix-Fayos (2011), Exploring particle size distribution and organic carbon pools mobilized by different erosion processes at the catchment scale, *J. Soils Sediments*, *11*, 667–678, doi:10.1007/s11368-011-0348-1.

Nadeu, E., A. A. Berhe, J. de Vente, and C. Boix-Fayos (2012), Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and land use change approach, *Biogeosciences*, *9*, 1099–1111, doi:10.5194/bg-9-1099-2012.

Nash, D. J., and S. J. McLaren (2003), Kalahari valley calcretes: their nature, origins, and environmental significance, *Quat. Int.*, *111*(1), 3–22, doi:10.1016/S1040-6182(03)00011-9.

Nelson, P. N., M. C. Dictor, and G. Soulas (1994), Availability of organic carbon in soluble and particle-size fractions from a soil profile, *Soil Biol. Biochem.*, *26*, 1549–1555, doi:10.1016/0038-0717(94)90097-3.

Okin, G. S., N. Mahowald, O. A. Chadwick, and P. Artaxo (2004), Impact of desert dust on the biogeochemistry of phosphorus in terrestrial ecosystems, *Glob. Biogeochem. Cycles*, *18*(2), doi:10.1029/2003GB002145.

Olk, D. C., K. G. Cassman, and T. W. M. Fan (1995), Characterization of two humic acid fractions from a calcareous vermiculitic soil: implications for the humification process, *Geoderma*, *65*(3–4), 195–208, doi:10.1016/0016-7061(95)94048-9.

Owens, L. B., R. W. Malone, D. L. Hothem, G. C. Starr, and R. Lal (2002), Sediment carbon concentration and transport from small watersheds under various conservation tillage practices, *Soil Tillage Res.*, *67*, 65–73, doi:10.1016/S0167-1987(02)00031-4.

Oyonarte, C., A. Pérez-Pujalte, G. Delgado, R. Delgado, and G. Almendros (1994), Factors affecting soil organic matter turnover in a Mediterranean ecosystems from Sierra de Gador (Spain): An analytical approach, *Commun. Soil Sci. Plant Anal.*, *25*(11-12), 1929–1945, doi:10.1080/00103629409369164.

Pacala, S. et al. (2007), The North American carbon budget past and present, in *The First State of the Carbon Cycle Report (SOCCR): The North American Carbon Budget and Implications for the Global Carbon Cycle*, edited by A. W. King, L. Dilling, G. P. Zimmerman, D. M. Fairman, R. A. Houghton, G. Marland, A. Z. Rose, and T. J. Wilbanks, pp. 29–36, 167–170, National Oceanic and Atmospheric Administration, National Climatic Data Center, Asheville, NC, USA.

Palis, R. G., G. Okwach, C. W. Rose, and P. G. Saffigna (1990a), Soil erosion processes and nutrient loss. II. The effect of surface contact cover and erosion processes on enrichment ratio and nitrogen loss in eroded sediment, *Aust. J. Soil Res.*, *28*, 641–658.

Palis, R. G., G. Okwach, C. W. Rose, and P. G. Saffigna (1990b), Soil erosion processes and nutrient loss. I. The interpretation of enrichment ratio and nitrogen loss in runoff sediment, *Soil Res.*, *28*, 623–639, doi:10.1071/SR9900623.

Palis, R. G., H. Ghadiri, C. W. Rose, and P. G. Saffigna (1997), Soil erosion and nutrient loss. III. Changes in the enrichment ratio of total nitrogen and organic carbon under rainfall detachment and entrainment, *Aust. J. Soil Res.*, *35*, 891–905.

Parsons, A. J., A. D. Abrahams, and S.-H. Luk (1991), Size characteristics of sediment in interrill overland flow on a semiarid hillslope, southern Arizona, *Earth Surf. Process. Landf.*, *16*, 143–152, doi:10.1002/esp.3290160205.

Parsons, A. J., A. D. Abrahams, and J. Wainwright (1994), Rainsplash and erosion rates in an interrill area on semi-arid grassland, southern Arizona, *Catena*, *22*, 215–226, doi:10.1016/0341-8162(94)90003-5.

Parsons, A. J., J. Wainwright, A. D. Abrahams, and J. R. Simanton (1997), Distributed dynamic modelling of interrill overland flow, *Hydrol. Process.*, *11*, 1833–1859, doi:10.1002/(SICI)1099-1085(199711)11:14<1833::AID-HYP499>3.0.CO;2-7.

Petrie, M. D., S. L. Collins, D. S. Gutzler, and D. M. Moore (2014), Regional trends and local variability in monsoon precipitation in the northern Chihuahuan Desert, USA, *J. Arid Environ.*, *103*, 63–70, doi:10.1016/j.jaridenv.2014.01.005.

Petrie, M. D., S. L. Collins, A. M. Swann, P. L. McCall, and M. E. Litvak (2015), Grassland to shrubland state transitions enhance carbon sequestration in the northern Chihuahuan Desert, *Glob. Change Biol.*, *21*(3), 1226–1235, doi:10.1111/gcb.12743.

Polyakov, V. O., and R. Lal (2004a), Modeling soil organic matter dynamics as affected by soil water erosion, *Environ. Int.*, *30*, 547–556, doi:10.1016/j.envint.2003.10.011.

Polyakov, V. O., and R. Lal (2004b), Soil erosion and carbon dynamics under simulated rainfall, *Soil Sci.*, *169*, 590–599.

Poulter, B. et al. (2014), Contribution of semi-arid ecosystems to interannual variability of the global carbon cycle, *Nature*, *509*(7502), 600–603, doi:10.1038/nature13376.

Proffitt, A., and C. Rose (1991), Soil erosion processes.II. Settling velocity characteristics of eroded sediment, *Soil Res.*, *29*(5), 685–695.

Puttock, A. (2013), Vegetation Change and Water, Sediment and Carbon Dynamics in Semi-Arid Environments, University of Exeter.

Puttock, A., J. A. J. Dungait, R. Bol, E. R. Dixon, C. J. A. Macleod, and R. E. Brazier (2012), Stable carbon isotope analysis of fluvial sediment fluxes over two contrasting C4-C3 semi-arid vegetation transitions, *Rapid Commun. Mass Spectrom.*, *26*, 2386–2392, doi:10.1002/rcm.6257.

Puttock, A., C. J. A. Macleod, R. Bol, J. A. J. Dungait, and R. E. Brazier (2013), Changes in ecosystem structure, function and hydrological connectivity in semi-arid grass to woody vegetation transitions, *Earth Surf. Process. Landf.*, *38*(13), 1602–1611, doi:10.1002/esp.3455.

Puttock, A., J. A. J. Dungait, C. J. A. Macleod, R. Bol, and R. E. Brazier (2014), Woody plant encroachment accelerates erosion of previously stable organic carbon from dryland soils, *J. Geophys. Res. Biogeosciences*, 2014JG002635, doi:10.1002/2014JG002635.

Qi, F., C. Guoduong, and M. Masao (2001), The carbon cycle of sandy lands in China and its global significance, *Clim. Change*, *48*(4), 535–549, doi:10.1023/A:1005664307625.

Quinton, J., J. Davies, and E. Tipping (2014), Modelling soil carbon movement by erosion over large scales and long time periods, vol. 16, pp. EGU2014–2082, EGU, Vienna, Austria.

Quinton, J. N., J. A. Catt, G. A. Wood, and J. Steer (2006), Soil carbon losses by water erosion: Experimentation and modeling at field and national scales in the UK, *Agric. Ecosyst. Environ.*, *112*, 87–102, doi:10.1016/j.agee.2005.07.005.

Quiroga, A. R., D. E. Buschiazzo, and N. Peinemann (1996), Soil organic matter particle size fractions in soils of the semiarid Argentinian Pampas, *Soil Sci.*, *161*, 104–108.

Ravi, S., P. D'Odorico, T. M. Zobeck, T. M. Over, and S. L. Collins (2007), Feedbacks between fires and wind erosion in heterogeneous arid lands, *J. Geophys. Res. Biogeosciences*, *112*(G4), n/a–n/a, doi:10.1029/2007JG000474.

Ravi, S., D. D. Breshears, T. E. Huxman, and P. D'Odorico (2010), Land degradation in drylands: Interactions among hydrologic–aeolian erosion and vegetation dynamics, *Geomorphology*, *116*, 236–245, doi:10.1016/j.geomorph.2009.11.023.

Rawling, G. C. (2005), *Geology and hydrologic setting of springs and seeps on the Sevilleta National Wildlife Refuge*, Open-file Report, New Mexico Bureau of Geology and Mineral Resources, New Mexico Tech, Socorro, New Mexico.

R Core Team (2015), *R: A language and environment for statistical computing*, R Foundation for Statistical Computing, Vienna, Austria.

Reynolds, J. F. et al. (2007), Global desertification: building a science for dryland development, *Science*, *316*, 847–851, doi:10.1126/science.1131634.

Rhoton, F. E., W. E. Emmerich, D. C. Goodrich, S. N. Miller, and D. S. McChesney (2006), Soil geomorphological characteristics of a semiarid watershed: influence on carbon distribution and transport, *Soil Sci. Soc. Am. J.*, *70*, 1532–1540, doi:10.2136/sssaj2005.0239.

Ridolfi, L., F. Laio, and P. D'Odorico (2008), Fertility island formation and evolution in dryland ecosystems, *Ecol. Soc.*, *13*(1), 13.

Ritchie, J. C. (1989), Carbon content of small reservoirs, *J. Am. Water Resour. Assoc.*, *25*, 301–308, doi:10.1111/j.1752-1688.1989.tb03065.x.

Robertson, G. P., and E. A. Paul (2000), Decomposition and soil organic matter dynamics, in *Methods in Ecosystem Science*, edited by O. E. Sala, R. B. Jackson, H. A. Mooney, and R. W. Howarth, pp. 104–116, Springer, New York, NY, USA.

Safriel, U. et al. (2005), Dryland Systems, in *Ecosystems and Human Well-being: Current State and Trends*, pp. 623–662.

Sainju, U. M., T. H. Terrill, S. Gelaye, and B. P. Singh (2003), Soil aggregation and carbon and nitrogen pools under rhizoma peanut and perennial weeds, *Soil Sci. Soc. Am. J.*, *67*(1), 146–155, doi:10.2136/sssaj2003.1460.

Sainju, U. M., W. F. Whitehead, and B. P. Singh (2011), Cover crops and nitrogen fertilization effects on soil aggregation and carbon and nitrogen pools, *Can. J. Soil Sci.*, doi:10.4141/S02-056.

Sankey, J. B., S. Ravi, C. S. A. Wallace, R. H. Webb, and T. E. Huxman (2012), Quantifying soil surface change in degraded drylands: Shrub encroachment and effects of fire and vegetation removal in a desert grassland, *J. Geophys. Res.*, *117*, G02025, doi:10.1029/2012JG002002.

Schiettecatte, W., D. Gabriels, W. M. Cornelis, and G. Hofman (2008a), Enrichment of organic carbon in sediment transport by interrill and rill erosion processes, *Soil Sci. Soc. Am. J.*, *72*, 50–55, doi:10.2136/sssaj2007.0201.

Schiettecatte, W., D. Gabriels, W. M. Cornelis, and G. Hofman (2008b), Impact of deposition on the enrichment of organic carbon in eroded sediment, *Catena*, *72*, 340–347, doi:10.1016/j.catena.2007.07.001.

Schlesinger, W. H., J. F. Reynolds, G. L. Cunningham, L. F. Huenneke, W. M. Jarrell, R. A. Virginia, and W. G. Whitford (1990), Biological feedbacks in global desertification, *Science*, *247*, 1043–1048, doi:10.1126/science.247.4946.1043.

Schlesinger, W. H., J. A. Raikes, A. E. Hartley, and A. F. Cross (1996), On the spatial pattern of soil nutrients in desert ecosystems, *Ecology*, *77*, 364–374, doi:10.2307/2265615.

Schlesinger, W. H., T. J. Ward, and J. P. Anderson (2000), Nutrient losses in runoff from grasslandand shrubland habitats in southern New Mexico: II. Field plots, *Biogeochemistry*, *49*, 69–86, doi:10.1023/A:1006246126915.

Scott, R. L., G. D. Jenerette, D. L. Potts, and T. E. Huxman (2009), Effects of seasonal drought on net carbon dioxide exchange from a woody-plant-encroached semiarid grassland, *J. Geophys. Res.*, *114*, G04004, doi:10.1029/2008JG000900.

Scott, R. L., J. A. Biederman, E. P. Hamerlynck, and G. A. Barron-Gafford (2016), The carbon balance pivot point of southwestern U.S. semiarid ecosystems: Insights from the 21st century drought, *J. Geophys. Res. Biogeosciences*, doi:10.1002/2015JG003181.

Sharpley, A. N. (1985), The selective erosion of plant nutrients in runoff, *Soil Sci. Soc. Am. J.*, *49*, 1527–1534, doi:10.2136/sssaj1985.03615995004900060039x.

Six, J., P. Callewaert, S. Lenders, S. De Gryze, S. J. Morris, E. G. Gregorich, E. A. Paul, and K. Paustian (2002), Measuring and understanding carbon storage in afforested soils by physical fractionation, *Soil Sci. Soc. Am. J.*, *66*, 1981–1987.

Slattery, M. C., and T. P. Burt (1997), Particle size characteristics of suspended sediment in hillslope runoff and stream flow, *Earth Surf. Process. Landf.*, *22*, 705–719, doi:10.1002/(sici)1096-9837(199708)22:8<705::aid-esp739>3.0.co;2-6.

Stallard, R. F. (1998), Terrestrial sedimentation and the carbon cycle: coupling weathering and erosion to carbon burial, *Glob. Biogeochem. Cycles*, *12*, 231–257.

Starr, G. C., R. Lal, R. Malone, D. Hothem, L. Owens, and J. Kimble (2000), Modeling soil carbon transported by water erosion processes, *Land Degrad. Dev.*, *11*, 83–91.

Starr, G. C., R. Lal, J. M. Kimble, and L. Owens (2001), Assessing the impact of erosion on soil organic carbon pools and fluxes, in *Assessment Methods for Soil Carbon*, edited by R. Lal, J. M. Kimble, R. F. Follett, and B. A. Stewart, pp. 417–426.

Stewart, J., A. J. Parsons, J. Wainwright, G. S. Okin, B. Bestelmeyer, E. L. Fredrickson, and W. H. Schlesinger (2014), Modelling emergent patterns of dynamic desert ecosystems, *Ecol. Monogr.*, *84*(3), 373–410, doi:10.1890/12-1253.1.

Throop, H. L., S. R. Archer, H. C. Monger, and S. Waltman (2012), When bulk density methods matter: Implications for estimating soil organic carbon pools in rocky soils, *J. Arid Environ.*, *77*, 66–71, doi:10.1016/j.jaridenv.2011.08.020.

Truman, C. C., T. C. Strickland, T. L. Potter, D. H. Franklin, D. D. Bosch, and C. W. Bednarz (2007), Variable rainfall intensity and tillage effects on runoff, sediment, and carbon losses from a loamy sand under simulated rainfall, *J. Environ. Qual.*, *36*(5), 1495–1502, doi:10.2134/jeq2006.0018.

Turnbull, L. (2008), Ecohydrological interactions across a semi-arid grassland to shrubland transition, University of Sheffield, Sheffield, UK.

Turnbull, L., J. Wainwright, and R. E. Brazier (2008a), A conceptual framework for understanding semi-arid land degradation: Ecohydrological interactions across multiple-space and time scales, *Ecohydrology*, *1*, 23–34, doi:10.1002/eco.4.

Turnbull, L., R. E. Brazier, J. Wainwright, L. Dixon, and R. Bol (2008b), Use of carbon isotope analysis to understand semi-arid erosion dynamics and long-term semi-arid land degradation, *Rapid Commun. Mass Spectrom.*, *22*, 1697–1702.

Turnbull, L., J. Wainwright, R. E. Brazier, and R. Bol (2010a), Biotic and abiotic changes in ecosystem structure over a shrub-encroachment gradient in the Southwestern USA, *Ecosystems*, *13*, 1239–1255, doi:10.1007/s10021-010-9384-8.

Turnbull, L., J. Wainwright, and R. E. Brazier (2010b), Changes in hydrology and erosion over a transition from grassland to shrubland, *Hydrol. Process.*, *24*, 393–414, doi:10.1002/hyp.7491.

Turnbull, L., J. Wainwright, and R. E. Brazier (2010c), Hydrology, erosion and nutrient transfers over a transition from semi-arid grassland to shrubland in the South-Western USA: A modelling assessment, *J. Hydrol.*, *388*, 258–272, doi:10.1016/j.jhydrol.2010.05.005.

Turnbull, L., J. Wainwright, and R. E. Brazier (2011), Nitrogen and phosphorus dynamics during runoff events over a transition from grassland to shrubland in the south-western United States, *Hydrol. Process.*, *25*, 1–17, doi:10.1002/hyp.7806.

Turnbull, L., A. J. Parsons, J. Wainwright, and J. P. Anderson (2013), Runoff responses to long-term rainfall variability in a shrub-dominated catchment, *J. Arid Environ.*, *91*, 88–94, doi:10.1016/j.jaridenv.2012.12.002.

Van Auken, O. W. (2009), Causes and consequences of woody plant encroachment into western North American grasslands, *J. Environ. Manage.*, *90*, 2931–2942, doi:10.1016/j.jenvman.2009.04.023.

Van Oost, K., G. Govers, T. A. Quine, G. Heckrath, J. E. Olesen, S. De Gryze, and R. Merckx (2005), Landscape-scale modeling of carbon cycling under the impact of soil redistribution: The role of tillage erosion, *Glob. Biogeochem. Cycles*, *19*, GB4014, doi:10.1029/2005GB002471.

Van Oost, K., J. Six, G. Govers, T. A. Quine, and S. De Gryze (2008), Reply to letter on "Soil Erosion: A carbon sink or source?" by R. Lal and D. Pimentel, *Science*, *319*, 1041–1042.

Voroney, R. P., J. A. Van Veen, and E. A. Paul (1981), Organic C dynamics in grassland soils. 2. Model validation and simulation of the long-term effects of cultivation and rainfall erosion, *Can. J. Soil Sci.*, *61*, 211–224.

Wainwright, J. (2005), Climate and climatological variations in the Jornada Experimental Range and neighbouring areas of the US Southwest, *Adv. Environ. Monit. Model.*, *2*.

Wainwright, J., and L. J. Bracken (2011), Runoff generation, overland flow and erosion on hillslopes, in *Arid Zone Geomorphology: Process, Form and Change in Drylands*, edited by D. S. G. Thomas, pp. 237–267, Wiley-Blackwell, Oxford, UK.

Wainwright, J., A. J. Parsons, and A. D. Abrahams (1995), A simulation study of the role of raindrop erosion in the formation of desert pavements, *Earth Surf. Process. Landf.*, *20*, 277–291.

Wainwright, J., A. J. Parsons, and A. D. Abrahams (1999), Field and computer simulation experiments on the formation of desert pavement, *Earth Surf. Process. Landf.*, *24*, 1025–1037.

Wainwright, J., A. J. Parsons, and A. D. Abrahams (2000), Plot-scale studies of vegetation, overland flow and erosion interactions: case studies from Arizona and New Mexico, *Hydrol. Process.*, *14*, 2921–2943.

Wainwright, J., A. J. Parsons, E. N. Müller, R. E. Brazier, D. M. Powell, and B. Fenti (2008), A transport-distance approach to scaling erosion rates: 1. Background and model development, *Earth Surf. Process. Landf.*, *33*, 813–826, doi:10.1002/esp.1624.

Wang, X., J. Wang, and J. Zhang (2012), Comparisons of three methods for organic and inorganic carbon in calcareous soils of Northwestern China, *PLoS ONE*, *7*(8), e44334, doi:10.1371/journal.pone.0044334.

Wang, X., L. H. Cammeraat, Z. Wang, J. Zhou, G. Govers, and K. Kalbitz (2013a), Stability of organic matter in soils of the Belgian Loess Belt upon erosion and deposition, *Eur. J. Soil Sci.*, *64*(2), 219–228, doi:10.1111/ejss.12018.

Wang, X., E. L. H. Cammeraat, P. Romeijn, and K. Kalbitz (2014a), Soil organic carbon redistribution by water erosion – the role of CO2 emissions for the carbon budget, *PLoS ONE*, *9*(5), doi:10.1371/journal.pone.0096299.

Wang, X., J. Wang, M. Xu, W. Zhang, T. Fan, and J. Zhang (2015), Carbon accumulation in arid croplands of northwest China: pedogenic carbonate exceeding organic carbon, *Sci. Rep.*, *5*, 11439, doi:10.1038/srep11439.

Wang, X. J., M. G. Xu, J. P. Wang, W. J. Zhang, X. Y. Yang, S. M. Huang, and H. Liu (2014b), Fertilization enhancing carbon sequestration as carbonate in arid cropland: assessments of long-term experiments in northern China, *Plant Soil*, *380*(1-2), 89–100, doi:10.1007/s11104-014-2077-x.

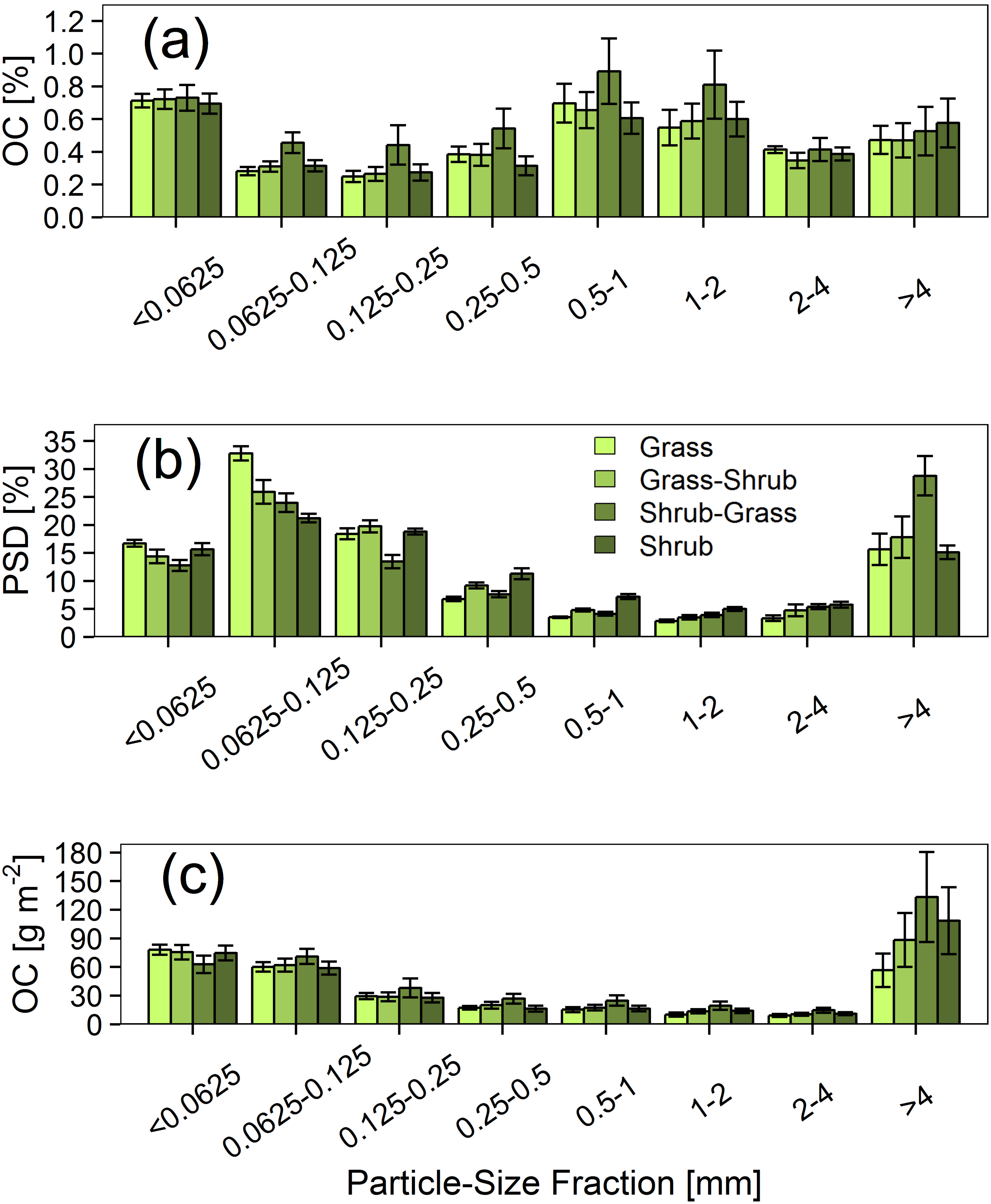
Wang, Z., G. Govers, A. Steegen, W. Clymans, A. Van den Putte, C. Langhans, R. Merckx, and K. Van Oost (2010), Catchment-scale carbon redistribution and delivery by water erosion in an intensively cultivated area, *Geomorphology*, *124*, 65–74, doi:10.1016/j.geomorph.2010.08.010.

Wang, Z., G. Govers, K. V. Oost, W. Clymans, A. V. den Putte, and R. Merckx (2013b), Soil organic carbon mobilization by interrill erosion: Insights from size fractions, *J. Geophys. Res. Earth Surf.*, *118*(2), 348–360, doi:10.1029/2012jf002430.

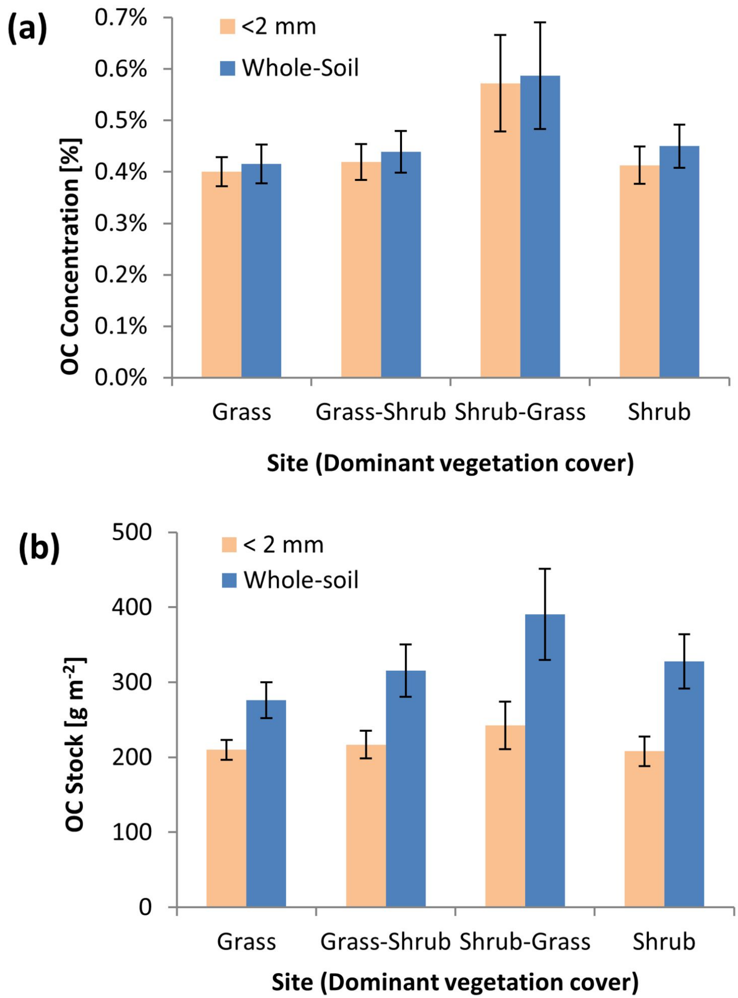
Wan, Y., and S. A. El-Swaify (1997), Flow-induced transport and enrichment of erosional sediment from a well-aggregated and uniformly-textured Oxisol, *Geoderma*, *75*, 251–265, doi:10.1016/S0016-7061(96)00093-6.

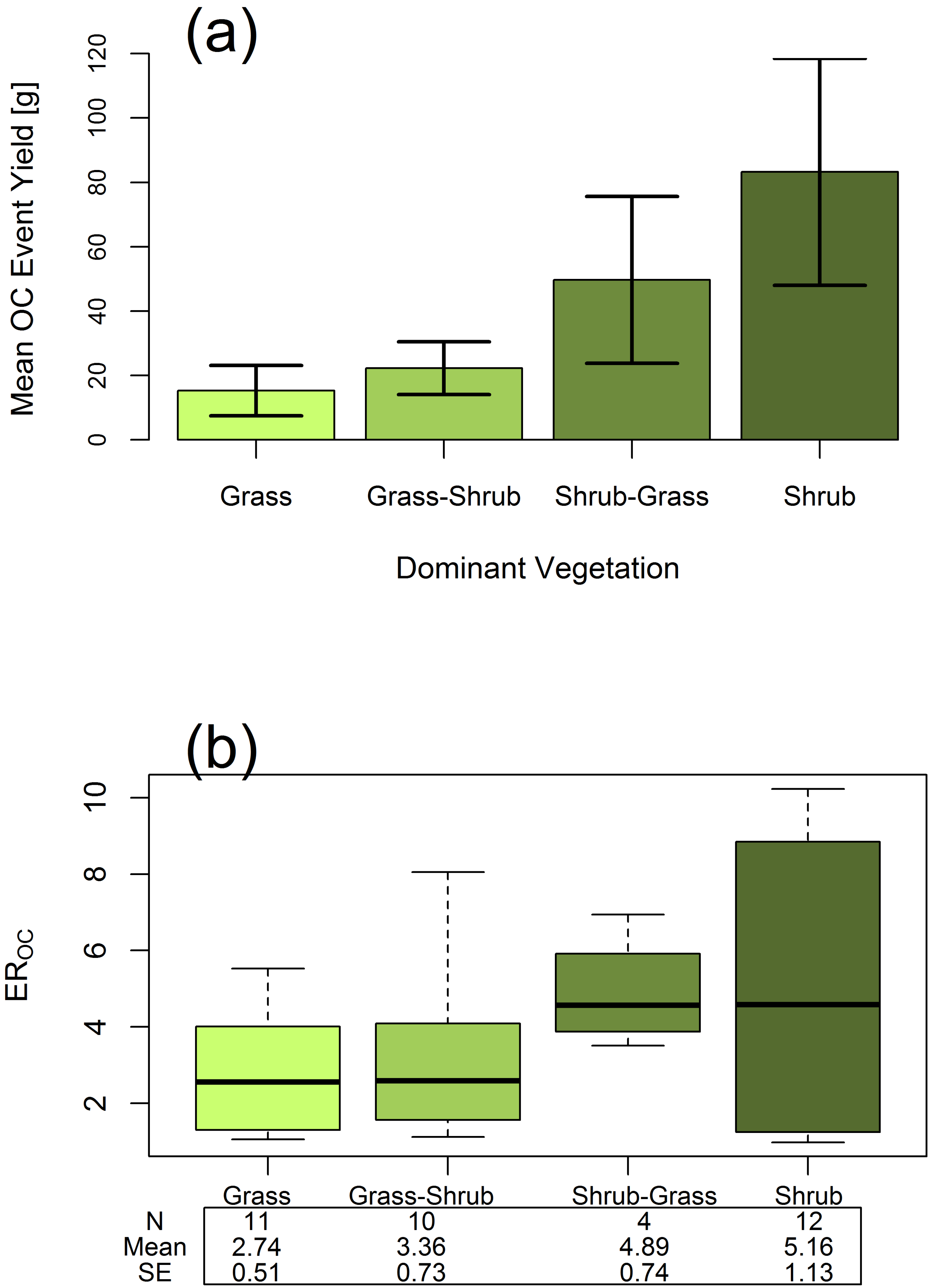
Wan, Y., and S. A. El-Swaify (1998), Sediment enrichment mechanisms of organic carbon and phosphorus in a well-aggregated oxisol, *J. Environ. Qual.*, *27*, 132–138, doi:10.2134/jeq1998.00472425002700010019x.

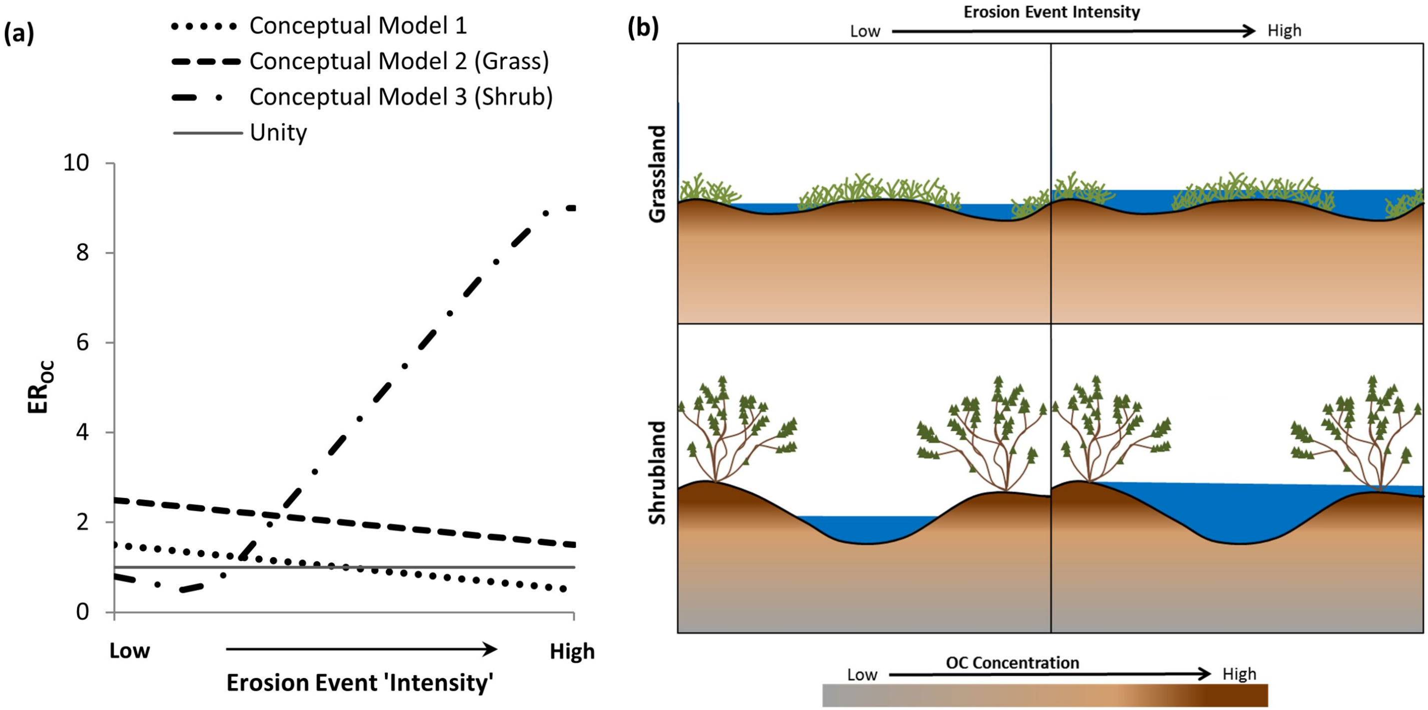
Wolkovich, E. M., D. A. Lipson, R. A. Virginia, K. L. Cottingham, and D. T. Bolger (2009), Grass invasion causes rapid increases in ecosystem carbon and nitrogen storage in a semiarid shrubland, *Glob. Change Biol.*, *16*, 1351–1365.



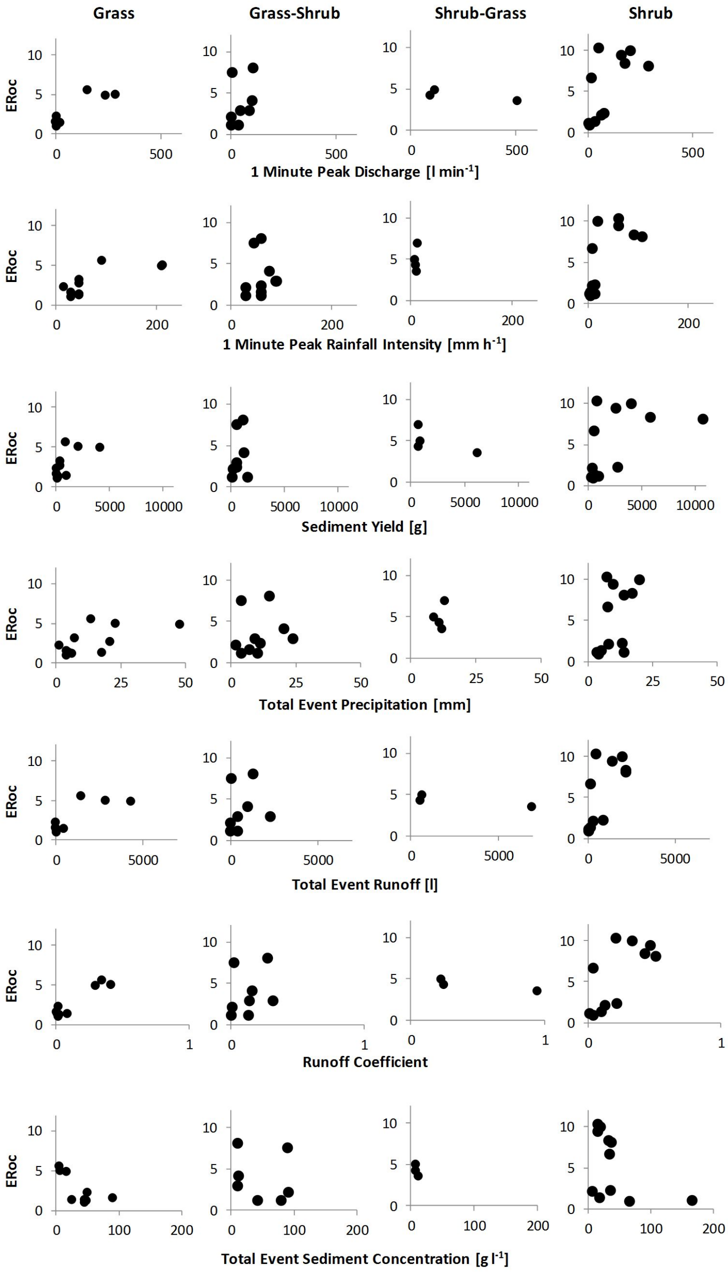
(a)
OC [%]
1.2
1.0
0.8
0.6
0.4
0.2
0.0
<0.0625
0.0625-0.125
0.125-0.25
0.25-0.5
0.5-1
1-2
2-4
>4
(b)
PSD [%]
35
30
25
20
15
10
5
0
Grass
Grass-Shrub
Shrub-Grass
Shrub
(c)
OC [g m-2]
180
150
120
90
60
30
0
Particle-Size Fraction [mm]


(a)
<2 mm
Whole-Soil
0.7%
0.6%
0.5%
0.4%
0.3%
0.2%
0.1%
0.0%
OC Concentration [%]
Grass
Grass-Shrub
Shrub-Grass
Shrub
Site (Dominant vegetation cover)
(b)
< 2 mm
Whole-soil
500
400
300
200
100
0
OC Stock [g m-2]
Grass
Grass-Shrub
Shrub-Grass
Shrub
Site (Dominant vegetation cover)


(a)
Mean OC Event Yield [g]
0
20
40
60
80
100
120
Grass
Grass-Shrub
Shrub-Grass
Shrub
Dominant Vegetation
(b)
ER_{OC}
2
4
6
8
10
Grass
Grass-Shrub
Shrub-Grass
Shrub
N 11 10 4 12
Mean 2.74 3.36 4.89 5.16
SE 0.51 0.73 0.74 1.13



(a)
Conceptual Model 1
Conceptual Model 2 (Grass)
Conceptual Model 3 (Shrub)
Unity
10
8
6
4
2
0
ER_{OC}
Low
High
Erosion Event 'Intensity'
(b)
Erosion Event Intensity
Low
High
Grassland
Shrubland
OC Concentration
Low
High


Grass
Grass-Shrub
Shrub-Grass
Shrub
ERoc
1 Minute Peak Discharge [l min-1]
1 Minute Peak Rainfall Intensity [mm h-1]
Sediment Yield [g]
Total Event Precipitation [mm]
Total Event Runoff [l]
Runoff Coefficient
Total Event Sediment Concentration [g l-1]