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**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

**Supporting Information:**

- Supporting Information S1

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# Dryland, calcareous soils store (and lose) significant quantities of near-surface organic carbon

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**Abstract** Semiarid 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 semiarid 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 that coarse fraction particles contain no OC are likely to underestimate soil OC stocks substantially, especially in soils with strongly stabilized aggregates. Sediment eroded from four hillslope scale (10 × 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. The average erosion-induced OC event yield increased sixfold 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 wildfire 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, among 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 characterize carbon stocks in semiarid 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 semiarid 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 semiarid 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 semiarid 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; *Z. Wang et al*., 2013, *Wang et al*., 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 modeling 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 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 homogenized repacked soils with synthetic structure, subjected to artificial rainfall [e.g., *Ghadiri and Rose*, 1991a, 1991b; *Palis et al*., 1990a, 1990b; *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; *X. Wang et al*., 2013], recent work has suggested that the enrichment of fine particles alone cannot explain observed OC enrichment [*Wang et al*., 2010; *Z. Wang et al*., 2013; *Chartier et al*., 2013].

Standard protocols for measuring soil organic carbon (SOC) discard the coarse (>2 mm) particle size fraction, assuming that 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; and (iv) to quantify differences in erosion-induced effluxes of OC across an ecotone from a grass-dominated to a shrub-dominated ecosystem over a 4 year period.

## 2. Methods

### 2.1. Study Site

The study site is located in the Mackenzie Flats of the Sevilleta National Wildlife Refuge in central New Mexico, USA (34°19′N, 106°42′W), experiencing a semiarid 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 analyzed 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*., 2010a, 2010b; *Puttock et al*., 2012, 2014; *Brazier et al*., 2013]. Thirty-seven 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 1 min 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, bed load traps, or natural sediment deposits risks being nonrepresentative 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 (Figure S1 and Table S1); for full description of the design and instrumentation of the plots, see *Turnbull et al*. [2010a, 2010b, 2011], *Puttock et al*. [2012, 2013, 2014], and *Brazier et al*. [2013].

### 2.3. Laboratory Analysis

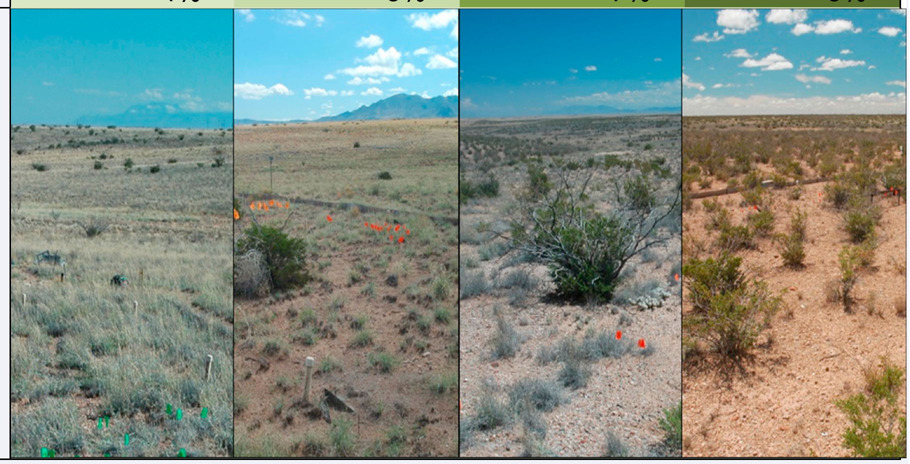
Investigations characterizing the chemistry of soil fractionated by particle size commonly deliberately disperse aggregates [e.g., *Quiroga et al*., 1996; *Six et al*., 2002; *von Lützow et al*., 2007; *Marzaioli et al*., 2010]. However, detailed investigations by *Chenu and Plante* [2006] have shown 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 deionized 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 $\varphi$ (Wentworth phi) intervals ($>4$, 4–2, 2–1, 1–0.5, 0.5–0.25, 0.25–0.125, 0.125–0.0625, and $<0.0625$ mm). Dry sieving was employed to minimize 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 min) disaggregates loosely aggregated particles, and increasing shaking duration (up to 30 min) showed no further changes in gravimetrically determined PSD, indicating that 10 min of shaking had disaggregated all loosely aggregated particles. This treatment helps to reduce problems with possible reaggregation 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 parameterize 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

**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][a]

| 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 | | | | |



[a]Photos by the author (July 2013). Note that column colours correspond to sites across the grass-shrub ecotone, and are also used in Figures 1 and 3.

fractionated by effective particle size into five size classes (>2, 2–0.5, 0.5–0.25, 0.25–0.0625, and <0.0625 mm). Relative to the eight size classes employed for the characterization 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. Five grams of each particle size fraction was digested in 75 mL of 2 *M* HCL for 7 days, filtered through a 0.45 μm filter, and triple rinsed with 100 mL of deionized 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 homogenized 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 analyzer (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 that aliquots were representative. In total, 592 unique samples were analyzed.

### 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 analyzed [*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, and 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

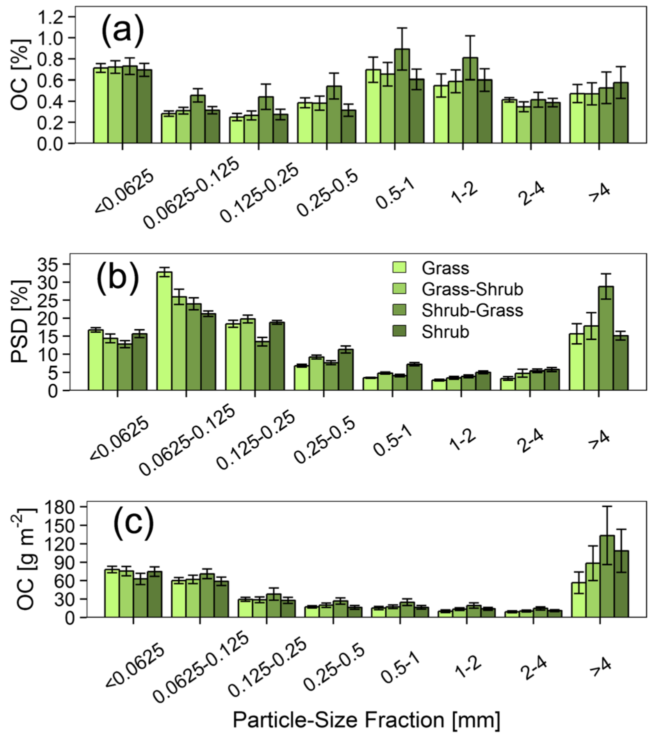



**Figure 1.** For each study plot across the grass-shrub ecotone: (a) areally weighted organic carbon (OC) concentrations observed in each particle size fraction, (b) areally weighted particle size distribution (PSD), and (c) areally weighted OC concentration in each particle size fraction in near-surface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

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 semiarid ecosystems exhibit high degrees of interevent 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) $\Sigma_{Obs}$ 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 that 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

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

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], where the total sediment concentration during each event ($C_{event}$) (g $L^{-1}$) was calculated as the total sediment yield ($S_{event}$) (g) normalized by the total runoff ($Q_{event}$) (L)

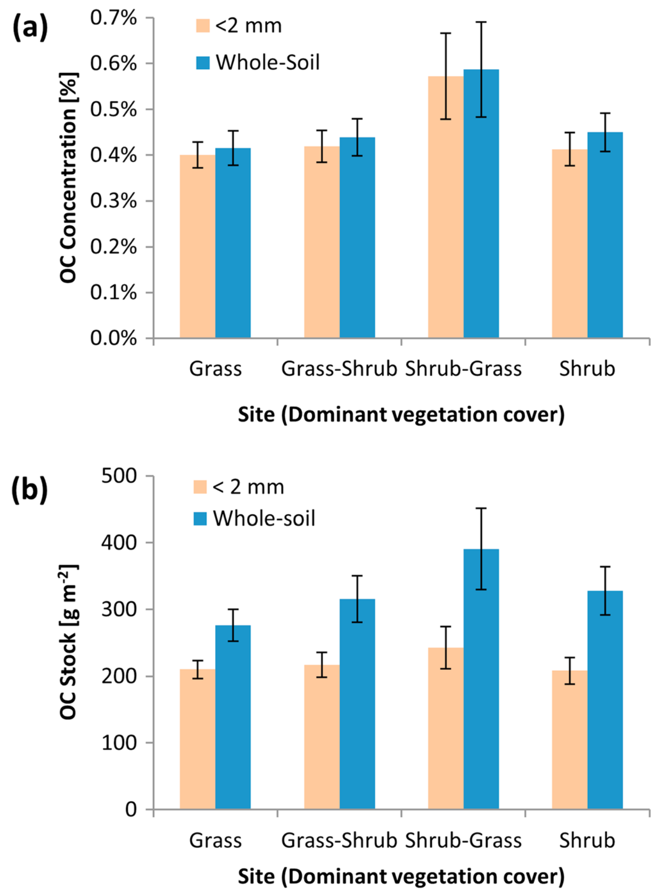



**Figure 2.** (a) Areally weighted near-surface (0–0.05 m) average organic carbon (OC) concentrations in the <2 mm fraction and whole soil. (b) Areally weighted near-surface soil organic carbon (OC) stocks for each site, calculated for the <2 mm fraction, and with the whole-soil OC concentration (including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

$$C_{event} = \frac{S_{event}}{Q_{event}} \quad (3)$$

Statistical analyses were conducted using R [*R Core Team*, 2015], and unless otherwise stated, all errors are standard errors (SEs). 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 analyzed to characterize 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 Figure 1a; this bimodal distribution was consistent in all of the average values for each surface cover type at all sites (data not shown). 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 1b). One hundred >2 mm aliquots were analyzed, 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 2a). These averages represent a wide range of concentrations and are not an artifact caused by the lower detection limit of the elemental analyzer.

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 2b). 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 1c). 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 (Figures 1b and 1c). Despite its relatively low OC concentration, the 0.125–0.0625 mm fraction contributes substantially (18% to 22%) toward the whole-soil SOC stock, primarily due to the abundance of particles in this size fraction (21% to 33% by weight) (Figures 1b and 1c).

### 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:

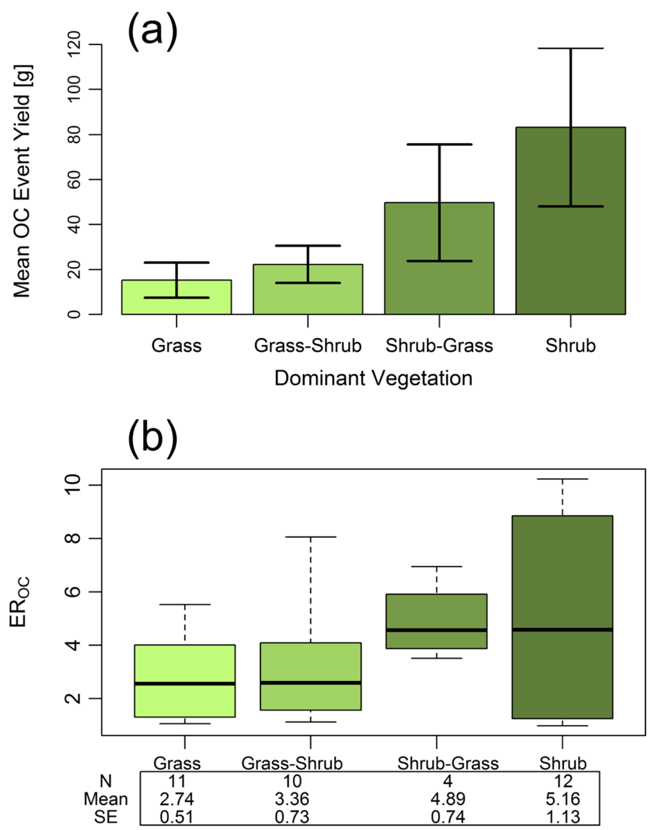



**Figure 3.** (a) Mean organic carbon (OC) event yield (± standard error) and (b) OC enrichment ratios and summary statistics, stratified by site. Where *N* is number of rainfall events, and SE is standard error. Bar colors correspond to sites across the grass-shrub ecotone (as shown in Table 1). In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

1. 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 kilometers of each other, the different plots therefore experienced different storm events over the monitoring periods [*Turnbull et al.*, 2010b].
2. 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 analyzed for OC yields ($n = 4$) (supporting information Figure S2). 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 sixfold 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 OC event yield between the two combined grass-dominated sites ($M$ = 18.65, SD = 25.57) versus the two combined shrub-dominated sites ($M$ = 74.87, SD = 108.26) was only statistically significant to the 6% level ($t$ = 2.034, df = 16.28, $p$ = 0.059). OC event yields were 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 3a). 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 3b). 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 3b). 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 that the difference in $ER_{OC}$ between the two amalgamated grass-dominated ($M$ = 3.04, SD = 2.07) and the two amalgamated shrub-dominated sites ($M$ = 5.09, SD = 3.42) was statistically significant ($t$ = 2.126, df = 23.17, $p$ = 0.044). 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

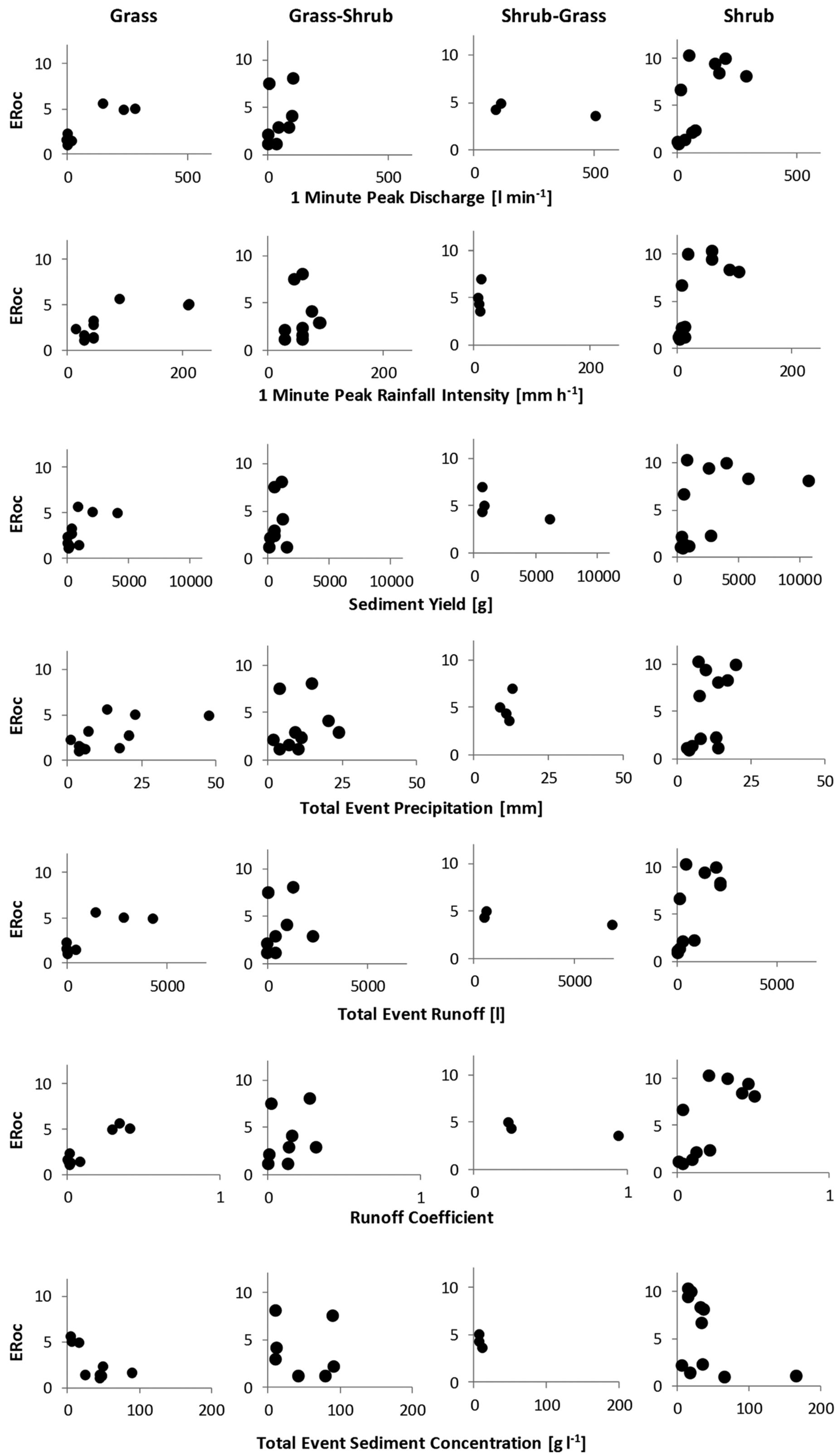



**Figure 4.** Relationships between the organic carbon enrichment ratio ($ER_{OC}$) and metrics of event intensity and magnitude: 1 min peak discharge, 1 min peak rainfall intensity, sediment yield, total event precipitation, total runoff, runoff coefficient, and total event sediment concentration ($C_{event}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

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, and 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 2a) and accounted for 24% to 38% of the total near-surface SOC stocks (Figure 1c). 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 that 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 (in the sense of *Agnelli et al*. [2002] and *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 $\varphi$ 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 reevaluation of the ubiquitous assumption that the coarse (>2 mm) fraction of the soil is free of OC, particularly in environments with stabilized 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 utilized 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 4 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 characterization 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 semiarid 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 help to elucidate emergent properties of resource redistribution processes in these ecosystems.

The sixfold 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] and *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], and ≤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; *X. Wang et al*., 2013]. 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 that 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, cause 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 toward 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 4 year study period, indicating that the previous finding may be an artifact 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 semiarid 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

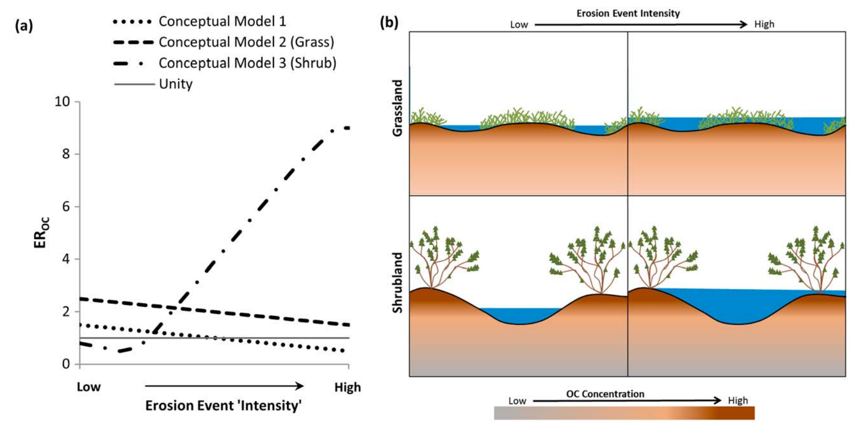



**Figure 5.** (a) Conceptual models of the relationship between event intensity and the enrichment ratio of organic carbon ($ER_{OC}$). Conceptual Model 1 reflects understanding from previous laboratory experiments documenting changes in sediment source areas (interrill versus rill) and associated degree 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, covarying 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.

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 10 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 5a. 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 5a). 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, nonhomogenized soils (Model 2 (Grass) in Figure 5a). 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 5b). Consequently eroded sediment may initially be depleted in OC ($ER_{OC} < 1$ in Figure 5a). As rainfall intensity increases, areas of topographic relief become inundated (Figure 5b) with greater erosion of material from these OC-rich areas, enhancing OC enrichment in the eroded sediment (Model 3 in Figure 5a). The variable source areas caused by covariation of topography and OC concentrations are hypothesized 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 interevent 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 min rainfall intensity, runoff coefficient, peak 1 min runoff, total runoff, total sediment event yield, and 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) 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 semiarid 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 that 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 semiarid environments, and there is a need for colocated 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 that 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 meaningful 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 6 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 semiarid shrubland but acknowledged that their findings did not quantify potential changes in erosional fluxes arising from the changes in vegetation structure. The $<1\,\text{g}\,\text{cm}^{-3}$ fraction of eroded material, 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 semiarid 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 sixfold 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.

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