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Use of rare earth oxides as tracers to identify sediment source areas for agricultural hillslopes

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Understanding sediment sources is essential to enable more effective targeting of in-field mitigation approaches to reduce diffuse pollution from agricultural land. In this paper we report on the application of rare earth element oxides to arable soils at hillslope scale in order to determine sediment source areas and their relative importance, using a non-intrusive method of surface spraying. Runoff, sediments and rare earth elements lost from four arable hillslope lengths at a site in the UK with clay soils were monitored from three rainfall events after tracer application. Measured erosion rates were low, reflecting the typical event conditions occurring at the site, and less than 1% of the applied REO tracers were recovered, which is consistent with the results of comparable studies. Tracer recovery at the base of the hillslope was able to indicate the relative importance of different hillslope sediment source areas, which were found to be consistent between events. The principal source of eroded sediments was the upslope area, implying that the wheel tracks were principally conduits for sediment transport, and not highly active sites of erosion. Mitigation treatments for sediment losses from arable hillslopes should therefore focus on methodologies for trapping mobile sediments within wheel track areas through increasing surface roughness or reducing the connectivity of sediment transport processes.

1 Introduction

Soil erosion from agricultural land is a major source of nutrients in freshwaters (Sharp-ley et al., 1994; Haygarth and Jarvis, 1999), contributing to eutrophication, and may cause increased turbidity and sedimentation in downstream waters (Owens et al., 2005). Sediment loss from arable land is of particular concern, as soil is easily eroded from bare and disturbed soil surfaces (Montgomery, 2007). Recent work (Withers et al., 2006; Deasy et al., 2009b; Li et al., 2007; Tullberg et al., 2007) has highlighted

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the role of tractor wheel tracks and trafficking in surface runoff erosion, but questions remain over whether wheel tracks act as source areas or are only transport pathways for sediment eroded from elsewhere in the field. This is because although sediment transfer can be easily observed and measured in runoff pathways, the detachment process itself is difficult to observe and quantify. Understanding the sources of sediment eroded in surface runoff, and their relative importance, is essential to provide data for the development and evaluation of soil erosion and sediment transport models, and enable more effective targeting of in-field mitigation approaches in order to reduce diffuse pollution from agricultural hillslopes.

Sediment tracers, which allow the movement of eroded soils to be tracked, offer a solution to the problem of determining sediment source areas. The most common field sediment tracing method is now sediment fingerprinting which relies on the inherent properties of the eroded and source material (Collins and Walling, 2004), while applied sediment tracer methods such as particle labeling of natural soil particles (Olmez et al., 1994) and mixing of magnetic tracers with soils (Ventura et al., 2001) have proved to be less applicable, principally because of the difficulties of applying tracers to large field areas. Because of the spatial variability of soil properties, sediment fingerprinting is an inappropriate technique where sediment sources and transport need to be defined at high spatial and temporal resolution. For artificial tracers, the main limitations are the differing properties of artificial tracers compared to natural material, and the lack of a variety of tracers to determine the spatial dynamics of soil erosion and sediment transport (Zhang et al., 2001). However, a number of recent studies have overcome these limitations through the use of rare earth oxide (REO) tracers. REOs are oxides available in powder form of the rare earth elements (REEs), the lanthanoid (formerly lanthanide) elements with atomic numbers 57 to 71. REOs display many of the properties required in a tracer (Zhang et al., 2001), being environmentally benign, and binding strongly to, but also being easily extracted from, the soil. Soils also contain low background concentrations of REEs, which means that large volumes of REO tracers are not required for tracing experiments, and in addition, REO tracers do not

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require heterogeneous soil physical and chemical properties within a catchment (Kimoto et al., 2006). One of the major benefits of using REOs as tracers is that different REO powders are available which can be applied to different soil areas and used as a spatial tool to trace the movement of sediment. Concentrations of REEs recovered in runoff can then be compared to concentrations of REEs applied as REO tracers and contained in background soil, and can be used to establish the sources and/or rates of erosion. A number of laboratory experiments have been undertaken to determine the applicability of REO tracers and understand the movement of sediment under simulated conditions (Zhang et al., 2001, 2003; Polyakov and Nearing, 2004; Lei et al., 2006; Liu et al., 2004). REOs have also been used in a limited number of studies to understand the movement of sediment in the field (Kimoto et al., 2006; Stevens and Quinton, 2008; Polyakov et al., 2004).

A number of different application methods have been used for previous REO tracer studies, including mixing REO powders with wetted soil and backfilling the soil area with the tracer and soil mix (Zhang et al., 2001), mixing REO powders with dry soil, undertaking repeated wetting and drying, then spreading on the soil surface and incorporating the mix into the soil (Polyakov et al., 2004), and mixing REO powders with dry soil then spreading the mix on the soil surface using sand to aid even spreading (Stevens and Quinton, 2008). Although direct mixing of REOs with soil has been shown not to substantially change the physicochemical properties of soil aggregates (Zhang et al., 2001), the mixing works best in dry soil (O. Pryce, personal communication, 2009), and backfilling is rarely appropriate outside the laboratory where areas to be tagged with tracer may be large. Spreading a REO and soil mix on the soil surface may change soil surface properties, particularly where soil pores and crusting have a role in runoff generation and sediment mobilization, and incorporation may be difficult at small scales or on no-till fields. An easy, non-intrusive application method is therefore essential if REO tracing techniques are to be used to understand sediment transport within large field scale experiments. This paper presents the results of an experimental study to determine the source areas and relative erosion rates for sediment eroded

from an arable hillslope in the UK using a novel non-intrusive REO tracer application method.

2 Experimental methods

The REO tracer experiment was undertaken on an arable farm at Loddington, Leicestershire, UK (Fig. 1). Four 1.5 m wide unbounded hillslope lengths were used for the tracer experiment. All hillslope lengths were cultivated up and down the slope with winter wheat (*Triticum aestivum* L.), two under minimum tillage, and two under traditional plough cultivation. Each of the hillslope lengths contained a tractor wheel track. Hillslope lengths were 69 m and 99 m, and were longer for the plough areas in order to incorporate all slope elements of concern. Four different REO tracers (Pr_6O_{11} , Nd_2O_3 , Sm_2O_3 and Gd_2O_3) were applied to four areas of interest for hillslope erosion, the top-slope, midslope, and downslope hillslope areas, and the wheel track areas (Fig. 1). Tracers were applied on 25 January 2008, and were banded across the hillslope so that all of the potentially connected hillslope area was tagged with the tracer.

The use of a tracer application method which did not disturb the soil surface was essential for this study as the tracer experiment took place during the third year of a field campaign exploring in-field mitigation options to reduce diffuse pollution losses from arable land to water (Deasy et al., 2009b). REO powders were applied to the hillslope in suspension in deionised water, using a manual knapsack sprayer (Allman X15, Allman Sprayers Ltd., Chichester, UK) with a constant pressure valve which allowed even flow rate. The concentration of the applied REO suspension varied depending on the REO, and the length of the hillslope to be sprayed. The approximate ground cover rate was 0.8 m s^{-1} , and the calibrated spray rate was 14 ml s^{-1} . The target application rate was 50 times background REE concentration assuming a 1 cm depth of interaction inferred from previous REO experiments in the laboratory (Stevens and Quinton, 2008). This target concentration was higher than that used in previously published studies, in order to provide greater difference between tagged and untagged areas.

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After REO tagging, surface runoff generated during rainfall events between January 2008 and April 2008 was collected in tanks at the base of each hillslope length. Further information on the experimental design, runoff measurement, water sample collection and analysis of suspended sediment in runoff is available in the literature (Deasy et al., 2009b). For the tracing experiment, sediment samples were collected from each hillslope length and used for determination of REE concentrations. Loads and yields of eroded sediment were calculated for each hillslope length.

Sediment samples were dried, lightly ground and sieved to <1 mm. Subsamples were then used for extraction of REEs using an adapted USEPA method (O. Pryce, personal communication, 2009), where 0.5 g samples were subjected to repeated heating using a heating block (SEAL BD50, SEAL Analytical, Fareham, Hampshire, UK) at 95°C with 20 ml HNO_3 and approximately 10 ml H_2O_2 . Samples were then diluted to 100 ml and filtered using $0.45\ \mu\text{m}$ Whatman filters before analysis. Extracted REE samples were analysed for Pr, Nd, Sm and Gd using ICP-OES (Varian 725-ES, Varian Ltd., Oxford, UK) or ICP-MS (Thermo Elemental X7, Fisher Scientific UK Ltd., Loughborough, UK) for the elements where samples were below ICP-OES detection limits ($0.01\text{--}0.03\ \text{mg l}^{-1}$). Internal standards were used for analysis. All extractions and analyses were also undertaken on blank samples and on external soil standards (NCS DC73319, China National Analysis Centre for Iron and Steel) for quality control.

Runoff (mm), sediment loads (kg), sediment yields (kg ha^{-1}), and REE loads (mg) for each hillslope length were calculated for each monitored rainfall event after tracer application. Depletion rates (%) or ratio of total mass of depletion of an element to mass of the applied element for a hillslope, and estimated erosion rates (kg ha^{-1}) using tracer proportions, were calculated for each hillslope area within tracer-tagged hillslope lengths (see Sect. S1 in Supplementary Material). Erosion rates were compared between different hillslope areas over time and for different cultivation treatments. Differences in erosion rates between treatments were analysed using GLM analysis in SPSS (PASW Statistics 17.0, SPSS Inc., Chicago, USA).

3 Results and discussion

3.1 Results

Three runoff events occurred in the winter and spring of 2008 after tagging of hillslope areas with REO tracers (Table 1). The three events monitored fit well within the range of rainfall events which occurred at Loddington in the study year (2007–2008), and in previous years (Table S1 and Fig. S2 in Supplementary Material), and can be considered to be representative of runoff occurring at the site under normal climatic conditions. The three rainfall events generated differing runoff responses (Fig. S3 in Supplementary Material). Event I was flashy in response to relatively high rainfall intensity. Event II had a long falling limb, in response to long duration moderate intensity rainfall. Event III was of low magnitude and long duration, as a result of lower rainfall intensities occurring over a longer period of time. Cumulative sediment losses from the four hillslope lengths used in the experiment averaged 20.0 kg ha^{-1} for all four hillslope lengths over the three events. Analysis of results from the three events indicates that there were no significant differences ($p < 0.05$) in sediment transport between the different soil cultivation types (Fig. S4, Supplementary Material), hence erosion source areas for the four hillslope lengths were considered together.

In the three events monitored, depletion rates for all hillslope lengths monitored and all hillslope areas were low (Fig. 2). The highest recovery rates over the three events for the applied tracers were measured for Gd (0.30%) and Nd (0.14%), hence over 99% of the applied tracer remained on the hillslope in the three events monitored.

Figure 3 illustrates clearly that the main source of sediment eroded from the hillslope lengths was sediment eroded from upslope areas (average for three events 69.0 kg ha^{-1}). Sediment eroded from within the wheel track areas averaged 12.1 kg ha^{-1} for the three events. Although SS yields in event II were significantly greater than in events I and III ($p < 0.05$), the relative importance of the different sediment source areas was similar over time. There were no significant differences in SS concentrations in runoff collected in the tanks between events.

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3.2 Discussion

3.2.1 Tracer depletion rates

Although tracer depletion rates for the Loddington hillslope appear to be very low (up to 0.4% over three events), these rates are consistent with REO recovery rates for a study in a small catchment in Ohio, USA, where depletion rates of only 0.83% to 8.19% were measured over 49 events (Kimoto et al., 2006). The low depletion rates are consistent with the low erosion rates generally observed for UK soils. For example, average annual UK erosion rates of around 600 kg ha⁻¹ for clay soils, 1500 kg ha⁻¹ for silt soils, and 2500 kg ha⁻¹ for sand soils are reported in the literature (Evans, 1996). Moreover, the low depletion rates are evidence to support the strong binding of REO tracers within hillslope soils demonstrated in laboratory studies (Zhang et al., 2001). A recent experiment (Polyakov et al., 2009) found that REO binding in the field was promoted by a number of rainfall events occurring after tagging of soils with the tracer, which did not generate runoff. Similarly, it is likely that the small rainfall event which occurred at Loddington in late January prior to event I, which was not enough to generate hillslope runoff (Fig. S2 in Supplementary Material), may have helped promote effective binding of the REO tracer after spraying, though it is possible some flushing of poorly incorporated REOs down the hillslope may have occurred in the first event.

Some of the applied tracer may also have been eroded and transported away from the hillslope via subsurface runoff pathways, for example through infiltration and field drainage. Recent research in artificially drained catchments, comparing surface and subsurface runoff pathways has demonstrated that subsurface runoff may account for a large proportion of the erosion occurring from a hillslope, particularly where surface runoff pathways largely correspond with tractor wheel tracks (Deasy et al., 2009a). Currently, there has been no consideration of this effect in the literature, due to the limited number of field studies undertaken using this novel tracer technique.

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3.2.2 Erosion source areas and processes

A previous study found that at the base of the Loddington hillslope, sediment could enter wheel tracks from unvegetated areas from as far away as 4 m (Stevens and Quinton, 2008). However, the results of this study suggest that sediment from the downslope area makes only a very small contribution to sediment eroded from the hillslope. The main source of sediment eroded from the hillslope in each event was the upslope area, implying that the wheel tracks, the main route of runoff and sediment transport within the arable hillslope, are principally conduits for hillslope erosion, and not highly active sites of erosion during the monitored events. This is consistent with the absence of observed rilling within the wheel tracks on the eroding plots during the study. The upslope area appears to act as a runoff generation zone, with water and sediment transferred to the wheeling channel via the connection of ponded areas. Although sediment eroded from the midslope and downslope areas would involve smaller transport distances, these areas do not appear to be as well-connected to the base of the slope as the upslope area, which was slightly shallower and more convex, and where wheel tracks may be less incised. Studies using ^{137}Cs as a tracer have shown that the severity of erosion, and hence erosion sources, can depend on slope shape, with greatest loss on convex slopes (Montgomery et al., 1997).

The cohesive clay soils at Loddington are also likely to be a factor in determining erosion sources. For example, in a watershed study on coarse gravelly soils in Arizona, channel contributions were the dominant erosion source (Polyakov et al., 2009), while in a similar watershed study in Ohio on silt loam soils, channel slope elements were the most important sediment source (Polyakov et al., 2004; Kimoto et al., 2006). In the silt loam watershed, it was also found that sediment eroded from a source area either moves only a short distance to adjacent areas, or is transported through the channel system to the base of the slope, which fits with the pattern of transport at Loddington.

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3.2.3 Tracer application method

Spraying tracers on to the soil surface in solution was an effective method for applying REO powders to soils. The calibrated sprayer resulted in even coverage of tracer over the soil surface, which could be seen visually (Fig. S1 in Supplementary Material). An even application rate was reliant on the ground coverage rate of the sprayer, but this was controlled by ensuring that all spraying was undertaken by the same user, and that up and downslope ground coverage rates were carefully timed. This application method is an improvement on the methods used in previous studies where soil and tracer mixes were broadcast, either by hand (Polyakov et al., 2004, 2009) or using a fertiliser spreader (Stevens and Quinton, 2008), being more accurate and allowing the solution to bind to the soil surface at a known rate which was low enough not to generate ponding, runoff or preferential infiltration through macropores. In agricultural catchments in previous studies, broadcast tracers were incorporated by repeated disk-ing and intense cultivation (Polyakov et al., 2004), which is likely to have influenced the high sediment yields measured in the first events, while others have avoided this problem by not incorporating tracers (Stevens and Quinton, 2008), which may have resulted in a layer of tagged material on the soil surface being readily available for entrainment. In both experiments, the necessary experimental preparation is likely to have influenced the results of the experiment. The surface spraying method presented here offers a workable technique which has a limited effect on the in-situ soil and allows the results of the experiment to represent naturally occurring erosion processes.

Previous studies exploring the use of REOs as sediment tracers at hillslope scale (Polyakov et al., 2004, 2009; Kimoto et al., 2006) have drawn attention to the issue of tracer dilution from untagged hillslope areas. This is particularly of concern where erosion depths occur which are greater than the tagging depth. However, on gently eroding hillslopes, such as those in the UK, where average erosion rates equate to soil lowering rates of around 0.4–1.8 mm yr⁻¹ (from Evans, 1996, assuming typical soil bulk density characteristics from Ruehlmann and Korschens, 2009), a tracer incorporation

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depth of 1 cm is likely to be appropriate and would allow sediment movement to be traced over periods of weeks to years. Where localized rilling and gullyng occur, then any REO tracer method where tracer is incorporated over whole hillslope areas may result in poor estimations of relative erosion rates, although it could still be used to indicate source areas.

3.2.4 Implications and future directions

The results from this UK field experiment suggest that REO tracers can be applied to large hillslope areas using surface spraying of powders in suspension, in order to trace source areas for sediments eroded from hillslopes in surface runoff. In the small erosion events which commonly occur in the UK, erosion depths are unlikely to exceed the depth of tracer interaction resulting from surface spraying, which means that dilution effects are unlikely to be a problem. Erosion rates at the Loddington field site were low, reflecting the typical event conditions occurring at the site, and little of the applied REO tracers were recovered, which is consistent with the results of comparable studies. Tracer recovery was however able to indicate the relative importance of different hillslope sediment source areas, which were found to be consistent between events.

Under typical event conditions, soil is principally eroded from gentler sloped more convex hillslope areas, which may not be closest to the base of the slope, where ponded areas are connected to preferential runoff pathways. For cohesive soils, erosion may not occur within the preferential pathways in all events, and sediment losses under typical event conditions are likely to be detachment limited, with sediment sources on arable hillslopes likely to be exhausted through a rainfall event. Mitigation treatments for sediment losses from arable hillslopes should therefore focus on methodologies for trapping mobile sediments within wheel track areas through increasing surface roughness or reducing runoff transport through ponding and infiltration.

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Supplementary material related to this article is available online at:
<http://www.solid-earth-discuss.net/2/195/2010/sed-2-195-2010-supplement.pdf>

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Event	Date	Rainfall (mm)	Peak Rainfall (mm hr ⁻¹)	Runoff Duration (hr)	Runoff (mm)	SS (mg l ⁻¹)	SS Yield (kg ha ⁻¹)
I	14 Feb 2008	25.6	19.2	95	0.7 (0.4)	1573 (1034)	13 (14)
II	19 Mar 2008	56	9.6	180	8.5 (7.2)	500 (283)	42 (33)
III	2 Apr 2008	21.8	9.6	227	0.8 (0.5)	893 (416)	5 (1)

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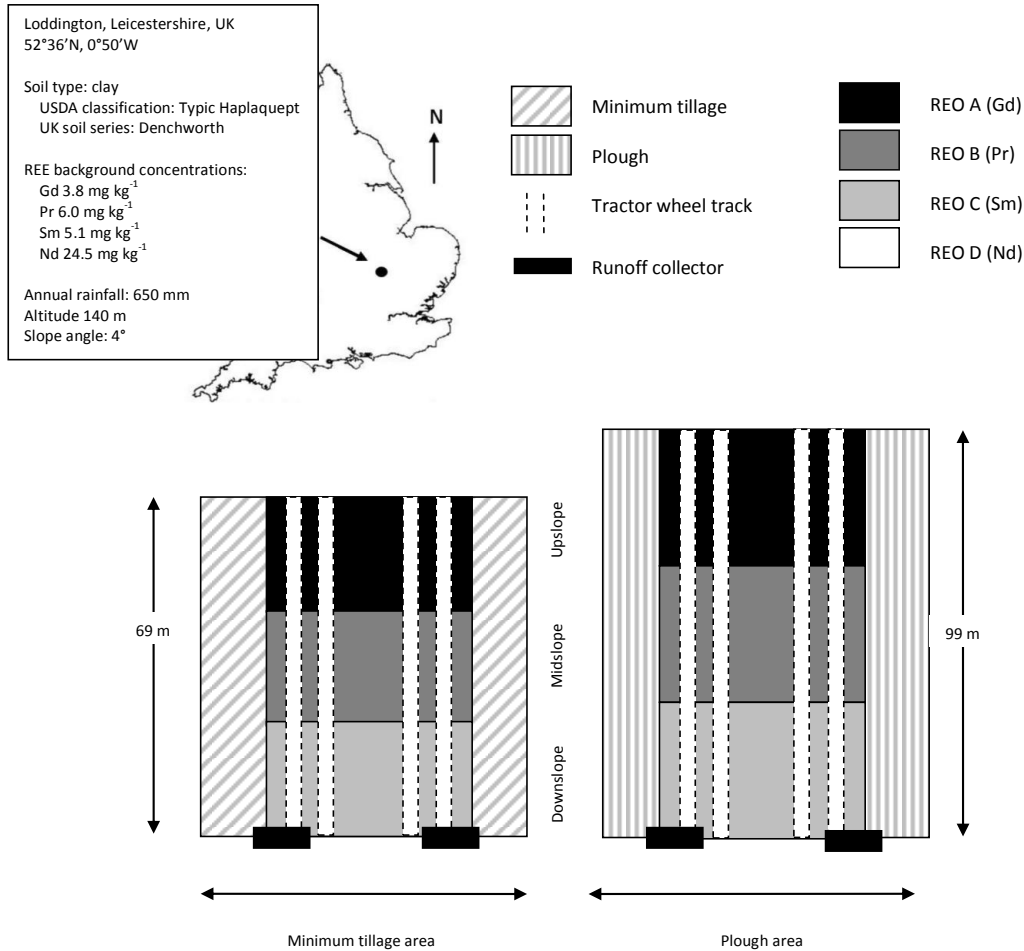


Fig. 1. Location of rare earth oxide (REO) tracer study site, background information and experimental design (not to scale).

Use of rare earth oxides as hillslope sediment tracers

C. Deasy and
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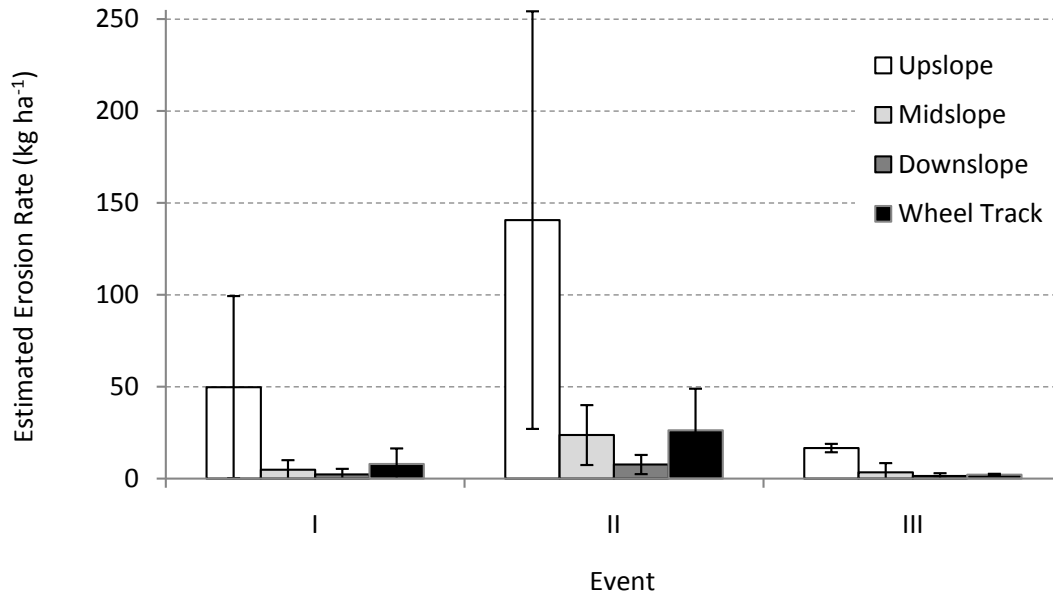


Fig. 3. Estimated erosion rates for hillslope areas tagged with rare earth oxides in three events at Loddington. Values are averages for four hillslope lengths, error bars are standard deviations.

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