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Variability in the mineral magnetic properties of soils and sediments within a single field in the Cape Fold mountains, South Africa: Implications for sediment source tracing

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ABSTRACT

When tracing the sources of sediment, reference to potential source groups is a necessity. However, the range of uncertainty present in source fingerprinting outputs has been shown to be increased by a high within-source group variability. The environmental factors controlling the magnetic properties of samples within a single field were examined on a particle size specific basis to continue building on the recent studies of the spatial variability associated with different tracer types. The study area was in the Eastern Cape of South Africa over a Quartzite geology and was covered by rough grassland.

Topsoils and gully walls were intensively sampled within the study field. Further samples were collected from weathering bedrock exposures, burnt soil and wetland soil in order to explore potential controlling factors on source magnetism. Each sample was fractionated to $125-63 \mu m$, $63-32 \mu m$, $32-25 \mu m$, $25-10 \mu m$ and $< 10 \mu m$.

Topographic position was a major controlling factor on soil magnetism, with saturated lowland gully walls and wetland topsoils being less magnetic than dry topsoils, especially within the $< 25 \ \mu m$ size fractions. The magnetism of the fields soils was primarily controlled by the concentrations of super paramagnetic and single domain grains formed through pedogenesis and combustion. The magnetism of samples increased with decreasing particle size. Dissolution of fine magnetic grains in the $< 25 \ \mu m$ fractions provided a potential basis for surface – subsurface source discrimination but also increased within-source group variability. The 25–10 μm fraction was a good compromise between good discrimination and a low within-source variability.

Little difference was found between the low frequency magnetic susceptibility of quartzite topsoils and the shale and sandstone topsoils found elsewhere in the Eastern Cape. However, igneous sources such as dolerite are far more magnetic than the quartzite topsoils. Discrimination between sediment sources can be highly variable within the < $63 \mu m$ fraction. For example, the < $10 \mu m$ fraction of soils from the study site had a higher magnetic susceptibility than Karoo shales (sieved to < $63 \mu m$), whilst the $63-32 \mu m$ fraction had a lower susceptibility than the Karoo shales.

1. Introduction

The properties (tracers) of sediment have been used to determine its provenance since the 1970s (Klages and Hsieh, 1975; Wall and Wilding, 1976), and recent years have seen the growing uptake and application of sediment source fingerprinting procedures (Walling, 2013; Miller et al., 2015; Walling and Foster, 2016; Walling and Collins, 2016; Collins et al., 2017). In conjunction with increased use of the source fingerprinting approach, a number of methodological developments have been tested and reported in the international literature. These include, amongst others, using new tracers such as those associated with sediment-associated carbon (Hancock and Revill, 2013), collection of different types of sediment samples in conjunction with the need to apportion sediment sources for informing management of different environmental issues such as spawning gravel siltation (Collins et al., 2013) or sediment-borne phosphorus transfers (Walling et al., 2008), and mass balance modelling for source ascription using either frequentist (Collins et al., 2010) or Bayesian (Stewart et al., 2014) approaches to uncertainty analyses.

Despite a range of methodological developments being reported for sediment source fingerprinting, an issue that continues to merit further appraisal concerns the spatial variability of tracer properties and the

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need to take explicit account thereof in the characterisation of individual sources included in any catchment sampling strategy (Collins and Walling, 2004). Early studies (e.g. Peart and Walling, 1986) apportioned sediment sources by averaging the source type tracer values to generate a single estimate for samples collected randomly across the study catchment. Here, it was argued that this averaging reflected the mobilisation and delivery of sediment from multiple locations to the river channel during storm events. The incorporation of uncertainty analyses in numerical source apportionment procedures subsequently resulted in either the mean or median of the tracer values for each source category being used in tandem with either parametric (e.g. standard deviation; Walling et al., 2008) or non-parametric (e.g. median absolute deviation or On: Collins et al., 2010) scaling statistics for generating distributions for repeat sampling by Monte Carlo analysis. The mixing models used by some studies were also structured to incorporate a weighting for spatial variability in source tracer values (Martinez-Carreras et al., 2008; Collins et al., 2010; Wilkinson et al., 2013). To improve the representativeness of random samples, some studies collected multiple replicates near individual sampling points for bulking into composites (Collins et al., 2010; Wilkinson et al., 2013). This approach tackles small scale spatial variability in tracers and permits the total sample numbers to remain viable in the context of the costs involved. Alternatively, some studies have adopted transect rather than spatially random source sampling strategies (Koiter et al., 2013). Regardless of whether a randomised or systematic approach is used, source samples must target actively eroding sources with the clear potential for connectivity to the river channel during effective rainfall events capable of mobilising and redistributing sediment. Connectivity may be in the form of narrow or breached riparian buffers between the source and the river channel or a clear transport pathway such as a ditch, road, farm track or gully.

Against this backdrop, some previous work has incorporated information on soil erosion estimated on the basis of either the RUSLE soil loss estimator (Renard et al., 1997) or fallout radionuclides (Wilkinson et al., 2015) to develop more representative source values for radiometric properties. Similarly, Du and Walling (2017) combined radiometric-based estimates of soil loss with spatial sampling of top soil in a small field to demonstrate an approach for generating erosion-weighted geochemical tracer values. Clearly, erosion rate will not be the only factor affecting tracer values for a given area or portion of a catchment and there remains a need to investigate the impact of additional controls on the spatial variability of widely used tracer properties.

Mineral magnetic properties were one of the earliest tracers utilised and have an extensive history of use (e.g. Walling et al., 1979; Thompson and Morton, 1979; Oldfield and Wu, 2000; Foster et al., 2008; Hatfield and Maher, 2009; Manjoro et al., 2017). Indeed, many studies have examined the magnetic properties of sediments in isolation without reference to potential sources present in the catchment (Foster et al., 2008; Hayashida et al., 2015). Whilst such approaches often yield valuable information on catchment processes, there are clear advantages to comparisons between sediment and source materials and this direct comparison underpins sediment source fingerprinting. The signature of sediment provenance is potentially complicated by numerous factors such as, particle size effects (Thompson and Morton, 1979; Oldfield et al., 1985), the in-growth of bacterial magnetite in deposited sediments (Li et al., 2009), within-source group variability (Blundell et al., 2009; Pulley et al., 2015b) and the post depositional dissolution of magnetic grains (Anderson and Rippey, 1988; Roberts and Turner, 1993; Foster et al., 1998). The collection and analysis of catchment source samples can aid in the interpretation of the complex magnetic signatures of sediment by acting as a frame of reference to identify the magnitude of these individual controlling factors for signature evolution from source to sink (Oldfield and Wu, 2000; Hatfield and Maher, 2009; Pulley et al., 2015a), as well as allowing for quantitative source apportionment (Walden et al., 1997).

Reference to potential catchment sediment sources requires the

categorisation of source groups (Walling et al., 1993; Collins and Walling, 2004). This is most commonly structured on the basis of land use (e.g. Peart and Walling, 1986) or geology (e.g. Owens et al., 1999). The range of uncertainty present in quantitative source fingerprinting outputs has been shown to be increased by a high within-source group variability in tracer concentrations and low contrasts in tracer signatures between different source groups (Small et al., 2002; Collins and Walling, 2002; Pulley et al., 2015). Within-source group variability can be increased by factors comparable to those causing magnetic tracer non-conservatism including, for example, the progression of rock weathering and soil formation (Torrent et al., 2010), the dissolution of magnetic grains (Grimley and Arruda, 2007), the in-growth of iron sulphides in saturated topsoils and subsoils (Staniek et al., 1994), fire (Clement et al., 2011), anthropogenic pollutant inputs (Shu et al., 2001), the selective export of fine particle sizes from a field (Quijano et al., 2014) and variations in local topography, geology and hydrological conditions (Blundell et al., 2009; Jordanova et al., 2012).

Sediment and soil particle size has been shown to exert a large effect on its magnetic properties (Thompson and Morton, 1979). As a result, changes to particle size during sediment erosion, transport and deposition can introduce significant uncertainty to a source tracing study. An additional consideration is that particle size may increase withinsource group variability, if there is spatial variability in the particle size distributions of soils and sediments. Importantly, discrimination between different sediment sources has been shown to be particle size specific (Hatfield and Maher, 2009; Pulley and Rowntree, 2016). For this reason, the fractionation of sediment source samples into narrow particle size ranges is becoming more common in tracing studies (Olley and Caitcheon, 2000; Hatfield and Maher, 2009; Pulley et al., 2015a; Laceby et al., 2017).

In South Africa, mineral magnetism has been extensively used to reconstruct historical sediment dynamics using both lake and floodplain deposits (Foster et al., 2007; Rowntree and Foster, 2012; Van der Waal et al., 2015; Pulley et al., 2015a; Manjoro et al., 2017; Mzuzu et al., 2016), creating the concomitant requirement for an understanding of the key controls on sediment source magnetism. Such work has been undertaken in the Karoo in the Eastern Cape by Pulley and Rowntree (2016), but is lacking in other regions of the country. This study therefore aimed to investigate the causes of spatial variability in mineral magnetic signatures within a single small field of a uniform quartzite geology and rough grassland land use in the Cape Fold mountains of South Africa. The environmental factors controlling magnetic properties of samples were examined on a particle size specific basis to determine how these different factors can affect the categorisation of a potential sediment source group, as well as the potential for using magnetic properties to differentiate between varied sources of sediment. An understanding of the effects of different environmental factors can guide sediment source tracing methods, and sample analysis such as the choice of particle size fraction and magnetic signatures to measure. This understanding can also contribute to the interpretation of the properties of sediment samples and their likely provenance. This targeted study was undertaken to continue building on the recent studies of spatial variability associated with radiometric and geochemical tracers described above, by focussing on mineral magnetic signatures.

2. Study site

The study field is located on the southern edge of Grahamstown in the Eastern Cape of South Africa. The geology of the area is composed of Quartzite of the Witpoort formation forming the eastern extent of the Cape Fold mountain range. The climate of the area is Mediterranean with an average annual rainfall of 683 mm, most which occurs in the summer months of October to March.

The study field (Fig. 1) is composed of a small valley with hills in the south and east. The field ranges in elevation between 620 and

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