



## Stages in the life of a magnetic grain: Sediment source discrimination, particle size effects and spatial variability in the South African Karoo



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### ABSTRACT

Mineral magnetic properties are commonly used to trace the sources of river and lake sediments with the aim of identifying areas of a catchment experiencing the high rates of soil loss. In some areas, however, there is poor understanding of how magnetic properties are able to discriminate between potential sediment sources. The basis for mineral magnetic source discrimination between topsoils and subsurface material, over different rock parent materials, was investigated in the South African Karoo. The effects of particle size, soil development/alteration and diagenesis processes on the source discrimination were investigated. Good discrimination (up to 100% correct discrimination) was found between the sedimentary sources (soils and subsurface) and dolerite soils due to much higher concentrations of magnetic minerals in the dolerite. There was little evidence of alterations to magnetic properties in subsurface sediments, resulting in only a small ability of properties to differentiate between surface and subsurface sources. It was found that during the development of topsoils there was a loss of high coercivity weakly magnetic grains and an enrichment in small superparamagnetic and stable single domain grains. Particle size was strongly related to magnetic properties with almost all of the secondary superparamagnetic and stable single domain grains created during soil formation being concentrated in the <32  $\mu\text{m}$  fraction and the loss of high coercivity grains taking place primarily in the >125  $\mu\text{m}$  fractions. The small ability of sediment to discriminate between surface and subsurface sources was based upon the dissolution of primarily  $\chi_{\text{fd}}$  and  $\chi_{\text{ARM}}$  carrying grains in the <32  $\mu\text{m}$  fractions and to a lesser extent the 63–32  $\mu\text{m}$  fraction. As a result it was recommended that source tracing with the <32  $\mu\text{m}$  fraction of soils and sediment should be treated separately from the 63–32  $\mu\text{m}$  and larger particle size fractions. The relationships between particle size and magnetic properties were fairly consistent in all soil samples in each source group, raising the possibility that reliable correction factors could be developed for each source group.

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### 1. Introduction

The use of tracers to determine sediment provenance has become a key tool in geomorphological research. Its major application is the investigation of the sources of fine sediment entering lakes and rivers, so that targeted mitigation measures can be applied to heavily eroding parts of a catchment (Walling, 2013). Source tracing exploits differences in tracer concentration between potential sediment sources to quantitatively or qualitatively apportion contributions of sediment from each source. Mineral magnetic properties have been extensively used to trace sediment sources in the South African Karoo (e.g. Foster et al., 2012; Rowntree and Foster, 2012; Van der Waal et al., in press) and worldwide (e.g. Oldfield, 1977; Maher et al., 2003; Zhang et al., 2008). A suite of magnetic measurements have the advantage over most tracers in that they are quick to measure, require relatively inexpensive equipment and are non-destructive.

Establishing a robust justification for the selection of tracers and establishing the basis for the ability of tracers to discriminate between sediment sources is an essential (Smith and Blake, 2014), but under examined part of the fingerprinting process. Magnetic properties have been shown to be able to discriminate between sediment sources on the basis of geology (Thompson and Oldfield, 1986), anthropogenic activity (Charlesworth et al., 2000), soil formation processes (Jordanova et al., 2011), and post-depositional alterations to deposited sediments and colluvium (Dillon and Bleil, 2006). During soil formation, magnetic properties are transformed by the conversion of amorphous or weakly magnetic iron minerals into strongly magnetic ferromagnetic grains (Le Borgne, 1955; Maher, 1984). This conversion has been shown to be primarily from large ferrimagnetic magnetites and maghemites (up to >110  $\mu\text{m}$  in diameter) to secondary antiferromagnetic grains (such as hematite) and fine grained (0.4–0.001  $\mu\text{m}$ ) magnetites and maghemites (superparamagnetic and stable single domain grains) (Maher, 1998; Sheng-Gao and Shi-Qing, 2008). The action of bacteria has been shown to be an important contributor to this magnetic enhancement of soils, with the rapid precipitation of fine superparamagnetic grains (Lovley et al., 1987). It has been suggested that this

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magnetic enhancement has varying effects on different particle size fractions, with reduced magnetic susceptibility in sand and coarse silt fractions and enhanced susceptibility in fine silt and clay fractions (Maher, 1998; Blake et al., 2006; Hatfield and Maher, 2009). Soil type and profile position have also been shown to affect its magnetic mineralogy, with the alteration of primary and secondary magnetic minerals by dissolution, authigenesis and diagenesis processes being able to cause differences between topsoils and subsurface soils (Singer and Fine, 1989) and among soils along a drainage transect (Grimley et al., 2004). While a variety of transformations to magnetic properties have been shown to take place, secondary magnetic minerals associated with soil formation can be absent in excessively wet or arid soils (Maher, 1998). In waterlogged soils iron sulphides may dominate a soil's magnetic properties (Stanjek et al., 1994) or the dissolution of magnetic minerals may take place (Grimley and Arruda, 2007). As with the magnetic enhancement of soils, dissolution has been shown to be particle size specific, preferentially affecting smaller grain sizes (Karlin and Levi, 1983) and is probably facilitated by iron-reducing bacteria (Kostka and Nealson, 1995). Sediments in long term storage show the opposite process to dissolution; the ingrowth of autochthonous bacterially produced iron oxides (Maher and Thompson, 1999; Oldfield and Wu, 2000). This is characterised by the production of stable single domain grains ( $<0.1 \mu\text{m}$ ), which are identified by a high  $\chi_{\text{ARM}}$  (Pan et al., 2005). High temperature combustion (such as in forest fires) has also been shown to increase the concentration of fine-grained ferrimagnets in soils (Mullins, 1977). It has been shown that ferrimagnetic mineral assemblages produced by fire have a finer grain size than those arising from weathering and soil formation alone (Oldfield and Crowther, 2007).

While magnetic properties have shown great potential for source tracing, uncertainties have been shown to be associated with their use. In some cases it is possible to correct for potential sources of error, in other cases correction methods still have to be developed. For example, there is a simple correction factor that can be applied to correct for the dilution effect of soil organic matter (Smith, 1999). As demonstrated above magnetic properties have also been shown to be sensitive to the particle size of a sample (see also Thompson and Morton, 1979). Unfortunately simple corrections for particle size as used for organic matter are of limited use because the relationships between particle size and magnetic properties have often been shown to be non-linear (Foster et al., 1998; Blake et al., 2006; Oldfield et al., 2009). Approaches aimed at accounting for complex particle size related uncertainties are uncommon in published literature. To reduce error many researchers have used narrow particle size bands for source tracing to limit particle size effects (Hatfield and Maher, 2009; Lacey and Olley, 2014). However, because magnetic enhancement or depletion is specific to different particle size fractions, by selecting narrow particle size bands for analysis we may overlook the potential for magnetic minerals formed through particle size sensitive processes to discriminate between sediment sources. Motha et al. (2003) measured the relationships between geochemical tracers and particle size and developed correction factors accordingly. There is, therefore, the potential for the mitigation of particle size effects when using mineral magnetic properties. However, Maher (1998) concluded that the enhancement of magnetic properties of paleosols must be assessed on a site specific basis due to the localised effects of dissolution and pedogenic enhancement. This means that if a high spatial variability exists, in the relationship between magnetic properties and particle size, then correction may not be possible.

The aim of this paper is to determine the reasons for magnetic discrimination between soils on different bedrock types as well as subsurface colluvium in the semi-arid Karoo of South Africa. These potential sediment sources are examined to determine the magnetic properties of the bedrock types and to gain insight in to how the formation of soils from the bedrock has resulted in the different magnetic mineralogies present. The effects of the subsurface storage of colluvium and alluvium

on magnetic properties is examined to determine whether processes of dissolution, authigenesis and diagenesis, have resulted in further changes to the magnetic properties of subsurface material. The effects of particle size on the magnetic properties is also investigated to determine the degree to which specific particle size fractions undergo different changes to their mineralogy during soil formation and subsurface storage. Our goal is to identify which particle size fractions are better able to discriminate between sediment sources. It was also evaluated whether relationships between particle size and magnetic properties vary on a site specific basis, to find out if the relationships are consistent enough to potentially produce reliable correction factors.

## 2. Study catchment

The study was conducted in the catchment of the Wilgerbosch River which drains the Sneeuwberg Mountains of the eastern Karoo, a semi-arid that receives an average of 423 mm annually (1908–2002 measured at Gordonville) (Grenfell et al., 2014). This rainfall is highly seasonal, with winter droughts and summer rainfall. The catchment lithology is composed of Jurassic Dolerites and Triassic Katberg Formation sandstone at high altitude, overlying Upper Permian to Triassic Balfour Formation shale that dominates lower slopes and valley floors. Thin and poorly developed soils are present on upper and lower slopes; the soils usually lack an A and sometimes a B horizon as a result of soil erosion (Boardman, 2014), with the bedrock often being exposed in the catchment. Headwater valleys in the Sneeuwberg area have been largely infilled with colluvium and alluvium to depths of up to ~8 m (Boardman et al., 2003). These valley bottom deposits are composed of a brownish colluvium and reddish palaeosol, which has been reported to be sometimes overlain by a grey organic layer attributed to past wet anaerobic conditions (Holmes et al., 2003). These colluvial deposits have been highly eroded locally, resulting in the formation of many badlands and gullies that may date back to the late nineteenth century, a time of stock grazing by European settlers (Rowntree, 2013). Stock grazing (domestic and game) persists as the predominant land use in the catchment. The vegetation is primarily composed of small shrubs and grasses, and bushfires have been recorded in the region (Manry and Knight, 1986).

## 3. Materials and methods

Sampling locations were distributed throughout the catchment as shown in Fig. 1. The sampling strategy was designed so as to collect samples from soils developed in the three main rock types (shale, dolerite and sandstone), including both topsoil and subsoil. Samples of topsoil were obtained from soils developed in shale (19), dolerite (14) and sandstone (10). A further 20 samples were collected from subsurface material. Two additional samples were collected from freshly weathering dolerite and shale bedrock (Fig. 2), to provide an indication of the magnetic properties of the freshly weathered rock.

Samples of topsoil (~2 kg) were collected from a depth of ~5–10 cm using a non-metallic trowel (locations shown in Fig. 1). Samples of subsurface material were collected from the exposed vertical faces of channel banks, gullies and badlands. The samples were collected from the entire channel bank or gully face apart from the top 10 cm of the soil profile. For both topsoil and subsoils, each sample comprised an amalgamation of 10 subsamples from within a 10 m radius of the sampling point, to increase the coverage of the sampling.

An ~200 g sub-sample was extracted from each source sample using a sample splitter. Prior to sieving the organic matter content of the samples was determined using loss on ignition at 450 °C with an ~10 g subsample of material (Grimshaw et al., 1989). The subsample was ultrasonically dispersed in a water bath containing distilled water for 10 min before being divided into seven particle size fractions by wet sieving. Particles greater than 2000  $\mu\text{m}$  in size were discarded before the samples were separated to 2000–1000  $\mu\text{m}$ , 1000–500  $\mu\text{m}$ ,

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