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Fingerprinting upland sediment sources: particle size-specific magnetic linkages between soils, lake sediments and suspended sediments

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ABSTRACT: Accelerated erosion of fine-grained sediment is an environmental problem of international dimensions. Erosion control strategies and targeting of mitigation measures require robust and quantitative identification of sediment sources. Here, we use magnetic 'fingerprinting' to characterize soils, and examine their affinity with and contribution to suspended sediments transported within two subcatchments feeding Bassenthwaite Lake, northwest England. A high-resolution soil magnetic susceptibility survey was made using a field susceptometer (ZH Instruments, SM400 probe). Combining the spatial and vertical (down-profile) soil magnetic data, a subset of soil profiles was selected for detailed, laboratory-based magnetic remanence analyses. The magnetic properties of the catchment soils are highly particle size-dependent. Magnetic analyses were performed on the 31–63 μm fraction, for particle size-specific comparison both with the suspended sediments and lake sediments. Fuzzy cluster analysis groups the soil magnetic data into six clusters, apparently reflecting variations in parent material and horizon type, with three magnetically hard soils as unclassified outliers. Examination of the cluster affinity of the soils, suspended sediments and lake sediments indicates that topsoils of the upper Newlands Valley and subsoils around Keskadale Beck are a major source of the Newlands Beck suspended load, and the recent (post-nineteenth century) sediments in the deep lake basin. Older lake sediments show strong affinity with a small number of the Derwent suspended sediments and one of the Glenderamackin soils. A large number of Derwent suspended sediments show no affinity with any of the soils or lake sediments, instead forming a coherent, discrete and statistically unclassified group, possibly resulting from mixing between the magnetically hard subsoils of the medium to high-altitude Glenderamackin and Troutbeck areas and softer, lower altitude Glenderamackin soils. The lack of any affinity of these suspended sediments with the lake sediments may indicate deposition along the Derwent flood plain and/or in the shallow delta of Lake Bassenthwaite. Particle size-specific magnetic fingerprinting is thus shown to be both highly discriminatory and quantitatively robust even within the homogeneous geological units of this catchment area. Such a methodological approach has important implications for small–large scale catchment management where sources of sediment arising from areas with uniform geology have been difficult to determine using other approaches, such as geochemical or radionuclide analyses. Copyright © 2009 John Wiley & Sons, Ltd.

KEYWORDS: environmental magnetism; magnetic susceptibility; sediment tracing; suspended sediment; soils; fuzzy clustering; English Lake District

Introduction

For catchments affected by accelerated erosion of fine particles, identification of point and diffuse sources of the eroding sediment is a key task. This is especially the case for upland areas, which often experience high vulnerability to soil erosion resulting from extremes in climate, relief and increased land-use pressure. Upland areas often provide niche habitats for many endangered and internationally protected species (Duigan, 2004). High biodiversity in upland areas results from the diverse nature of land cover, use and type. Within the temperate zone, upland areas account for a significant proportion of, for example, Western Europe. Upland temperate lakes subjected to accelerated fine sediment delivery (e.g. Dearing *et al.*, 1987; Ballantyne, 1991; van der Post *et al.*, 1997; Hatfield *et al.*, 2008) can be affected by poor water quality, eutrophication, reduced light penetration and clogging of spawning gravels (e.g. Winfield *et al.*, 2004; Owens *et al.*, 2005). Within upland catchments, degradation of sometimes thin, often nutrient-poor soils is another result of accelerated erosion. Fine sediment, an important vector for the transport of nutrients and pollutants, is dominantly transported in episodic pulses as suspended load in rivers. Identification of suspended sediment sources remains a key requirement in the targeting of sediment control strategies and mitigation measures aimed at reducing accelerated rates of erosion, in order to preserve biodiversity and improve ecological status in upland areas.

Sediment 'fingerprinting' is potentially the most direct and discriminatory method for identification of sediment sources in a range of international settings (e.g. Collins *et al.*, 1997a, 1998; Collins and Walling, 2002). Reflecting natural variations in radionuclide, geochemical and/or molecular properties, 'fingerprinting' of potential sources has enabled discrimination of diverse point and diffuse sources of sediment, including forest roads (Gruszowski, 2003; Minella *et al.*, 2008), topsoil (Collins *et al.*, 1997b; Walling *et al.*, 1999), arable land (Peart and Walling, 1986; Walling and Amos, 1999), land under pasture (Collins *et al.*, 1997a, 1997b), subsurface (Russell *et al.*, 2001; Walling *et al.*, 2008), channel banks (Walling *et al.*, 1979; Peart and Walling, 1988), landslides (Nelson and Booth, 2002) and urban sources (Carter *et al.*, 2003). Although the sophistication of quantitative techniques and parameter diversity has evolved rapidly in the last 10–15 years, many studies still rely on one-dimensional, concentration-related sediment variables, rendering discrimination within relatively homogeneous catchments difficult. More complex methods of sediment characterization are both time- and cost-intensive which may account for their slow uptake and implementation. Conversely, a typical suite of room temperature magnetic measurements can rapidly and non-destructively discriminate a sample in several dimensions (e.g. by magnetic concentration, mineralogy and grain size) and be highly sensitive to subtle changes in a range of environmental settings (e.g. Thompson *et al.*, 1975; Maher, 1986, 1998). In sediment sourcing contexts, use of discriminatory and conservative magnetic properties has enabled matching of potential soil sources to the suspended load (e.g. Walling *et al.*, 1979), discrimination of suspended load contributions (e.g. Caitcheon, 1993; Hatfield and Maher, 2008; Maher *et al.*, 2009) and construction of millennial records of environmental history and change (e.g. Dearing, 1999; Dearing *et al.*, 2001; Oldfield *et al.*, 2003; Hatfield and Maher, 2009). Magnetic properties can be highly particle size-dependant (e.g. Oldfield *et al.*, 1985; Foster *et al.*, 1998; Hatfield and Maher, 2008). Thus their resolving power can be increased through measurement of specific clastic ranges, whilst the use of statistical 'unmixing' models enables quantitative source ascription (desirable to policy-makers and environmental managers). Considerable as yet unused potential exists for quantitative provenancing of suspended sediment by applying magnetic measurements on a particle size-specific basis.

Bassenthwaite Lake, an upland area in the English Lake District and a site of special scientific interest (SSSI), provides one of only two natural UK habitats for the rare and threatened whitefish, the vendace (*Coregonus albuda*). The lake currently suffers problems arising from accelerated fine sediment delivery. Vendace numbers are thought to be in decline due to fine sediment clogging their spawning gravels and increasing lake nutrient loading. The 240 km^2 catchment of Bassenthwaite Lake contains three major river systems, the Greta/Derwent/Glenderamackin (GGD), Newlands Beck, and Chapel Beck; and three lakes, Bassenthwaite, Derwent Water and Thirlmere (Figure 1a). The catchment geology is dominated by the Skiddaw Slate Group (SSG) though the south-eastern portion of the catchment lies within the Borrowdale Volcanic Group (Figure 1b). Within the SSG, the Kirkstile Formation (laminated mudstone and siltstone with greywacke sandstones) is dominant in the north and the Buttermere Formation (sheared and folded mudstone, siltstone and sandstone) is dominant in the south-western Newlands Valley (BGS, 1999). The Newlands Valley subcatchment is more 'mountainous' and hydrologically flashy compared to the more open and rolling GGD subcatchment, possibly reflecting both the differences in geology and the Newlands' more northerly aspect. The distribution of soil types within the catchment is heavily influenced by altitude and slope. Blanket peat occupies the mountainous plateau areas; shallow acid peaty soils dominate the upper slopes; deeper soils, the lower slopes. Valley bottoms suffer seasonal waterlogging through slowly permeable clayey soils and considerable thicknesses of river alluvium have been deposited between Derwent Water and Bassenthwaite Lake (Figure 1c). Present day land use is dominated by pastoral farming in the valley bottoms, concentrated in riparian areas whilst historical land use has included significant mining of lead, copper and baryte, initiated as late as the early nineteenth century in the previously unexploited Newlands ('new lands') Valley with the resulting catchment disturbance responsible for increased sediment fluxes during peak yields (Hatfield *et al.*, 2008; Hatfield and Maher, 2009). In more recent times land-use and climate appear to be more influential on the sedimentary regime in Bassenthwaite (Hatfield *et al.*, 2008; Hatfield and Maher, 2009).

Particle size-specific magnetic analysis of contemporary suspended sediments has enabled discrimination between the three main subcatchment inflows to Bassenthwaite, the GGD system, Newlands Beck and Chapel Beck, and authigenic, in-lake sources of magnetite (Figure 2a; Hatfield and Maher, 2008; Hatfield *et al.*, 2008). Comparison of the inflow suspended sediment loads with the Bassenthwaite Lake sediments indicates that the GGD was the major sediment source to Bassenthwaite for much of the mid-late Holocene (Figure 2b; Hatfield and Maher, 2009). In contrast, for the last 300 years, Newlands Beck appears to have been the dominant sediment source to the deep lake basin, with unprecedented increases in sediment flux within the last 100 years (Figure 2b; Hatfield and Maher, 2009; Hatfield *et al.*, 2008). Here, we build upon this magnetic 'fingerprinting' of contemporary suspended sediments and modern and older lake sediments in the Lake Bassenthwaite catchment, and attempt particle size-specific magnetic discrimination of the source soils contributing to accelerated sediment flux at the present day and in the past.

Methods

Field measurements

Catchment surveys of magnetic susceptibility have proven useful for identification of, and delineation between, different soil units (Williams and Cooper, 1990; Lees, 1994; Dearing *et al.*, 1996, Grimley *et al.*, 2004). However, such surveys have until recently mostly been restricted to surface sensing of soil magnetic properties. Here, for the Bassenthwaite catchment, a ZH Instruments SM400 magnetic susceptibility profiler and HUMAX soil corer were used in combination, to reduce sampling time to minutes per site, enabling catchment-wide magnetic survey of *in situ* soil profiles. Spatial resolution varied between <1 km^2 and ~3 km^2 in the Newlands Valley, up to ~9 km^2 in the GGD. In total, 64 sites were measured and sampled. The HUMAX corer removes a soil core (retained for subsequent laboratory analysis) from the sampling site. The SM400 susceptibility profiler, inserted into the bored hole, provides high-resolution (up to six measurements per millimetre), quasi-continuous measurement of *in situ*, down-profile magnetic susceptibility (Petrovsky *et al.*, 2004). Sampling sites were selected to encompass environmental variations in soil type and geology and also physical characteristics e.g. slope and drainage, all of which may influence soil magnetic properties (e.g. Maher, 1998). Driven by hand and equipped with plastic core sleeves, the HUMAX corer retrieved soil

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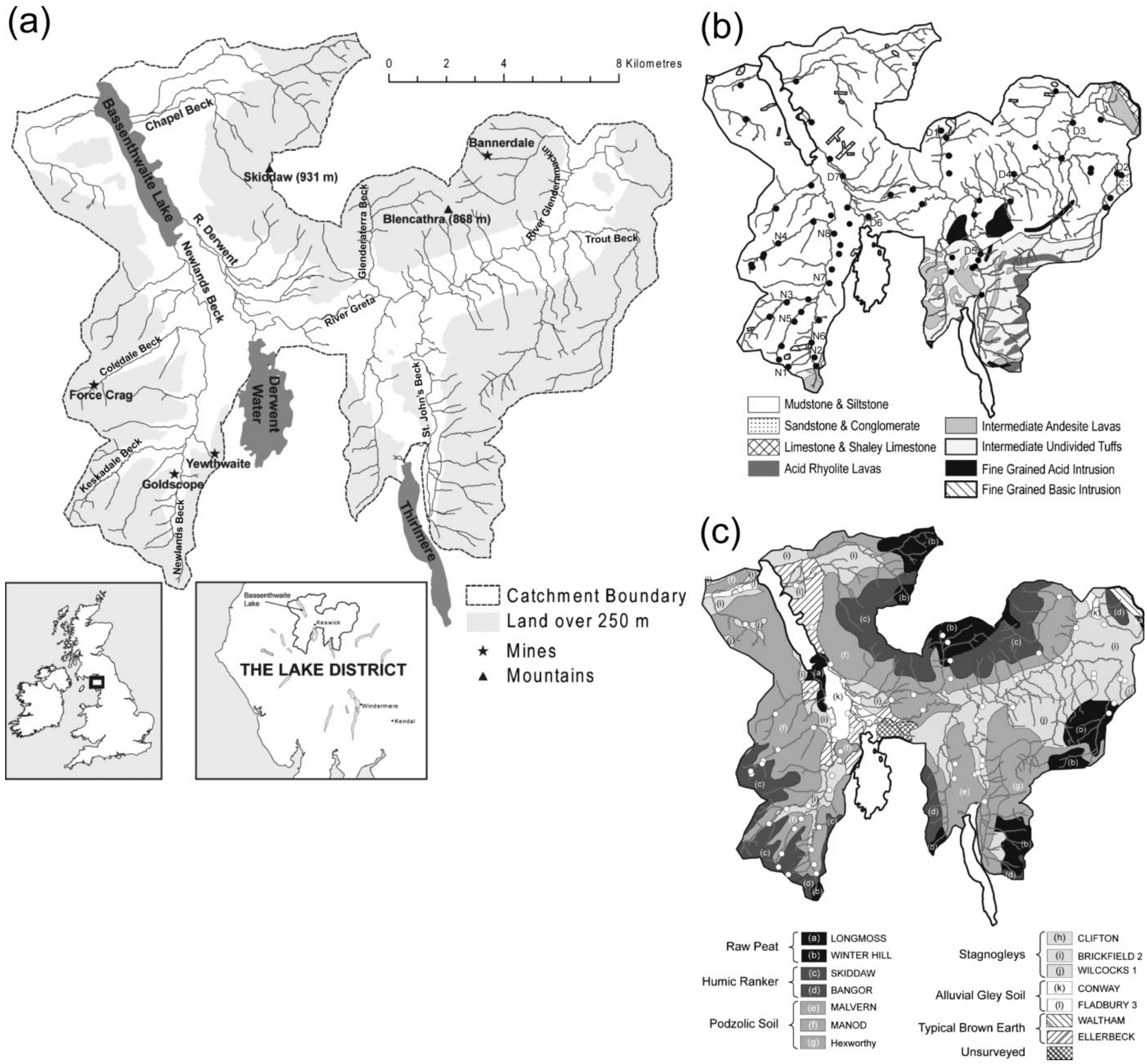

Figure 1. The Bassenthwaite catchment showing: (a) main subcatchments, tributaries, lakes, relict mining locations and land above 250 m; (b) geological variations, with locations of the 64 soil cores and 15 selected profiles; (c) soil types, adapted from the National Soil Survey of Great Britain.

cores up to ~30 cm long (soil compression resulting in significant core shortening), sampling both topsoil and subsoil in these relatively thin upland soils. Upon return to the laboratory, cores were stored in a refrigerator at 4 °C to prevent post-recovery transformation of material. Sample locations were recorded using a hand-held global positioning system (GPS) unit with a typical accuracy <10 m.

Laboratory analyses

In the laboratory, the soil cores were measured undisturbed in their plastic sleeves at 5 mm intervals using a Bartington MS2 susceptibility scanner with a 62 mm core loop (C) attachment. Cores were then removed from their sleeves, photographed and subjected to standardized soil profile descriptions which included measurement of colour, texture, and pH (using a digital probe accurate to 0·1 pH units). A subset of 15 cores was then selected. These soils were sectioned at 1 cm intervals, weighed and immobilized in 10 cc plastic pots and their low frequency (0·46 kHz) and high frequency (4·6 kHz) magnetic susceptibility measured, using a Bartington MS2B single sample sensor (sensitivity 10^{-7} SI units). The frequency dependence of susceptibility (χfd %), given as $\{[(\chi lf - \chi hf)/\chi lf] \times 100\}$, is proportional to the concentration of ultrafine magnetic grains around the stable single domain/superparamagnetic (SSD/SP) boundary (~0·03 μm in magnetite, for example). Samples were dried overnight at 40 °C, re-weighed and susceptibility re-measured to identify any magnetic loss upon drying/oxidation (e.g. in the case of presence of magnetic iron sulphides, such as greigite). For each of the 15 cores, a representative topsoil and subsoil sample were selected for particle size separation into five size fractions. To optimize particle sizing, 25 ml Calgon solution (10%) was added to samples prior to ultrasonic dispersion for 15 minutes. The >63 μm fraction was separated by wet sieving; the 31–63, 8–31, 2–8 and <2 μm fractions were separated by timed settling in Atterberg columns. The separated particle size fractions were dried at 40 °C, weighed

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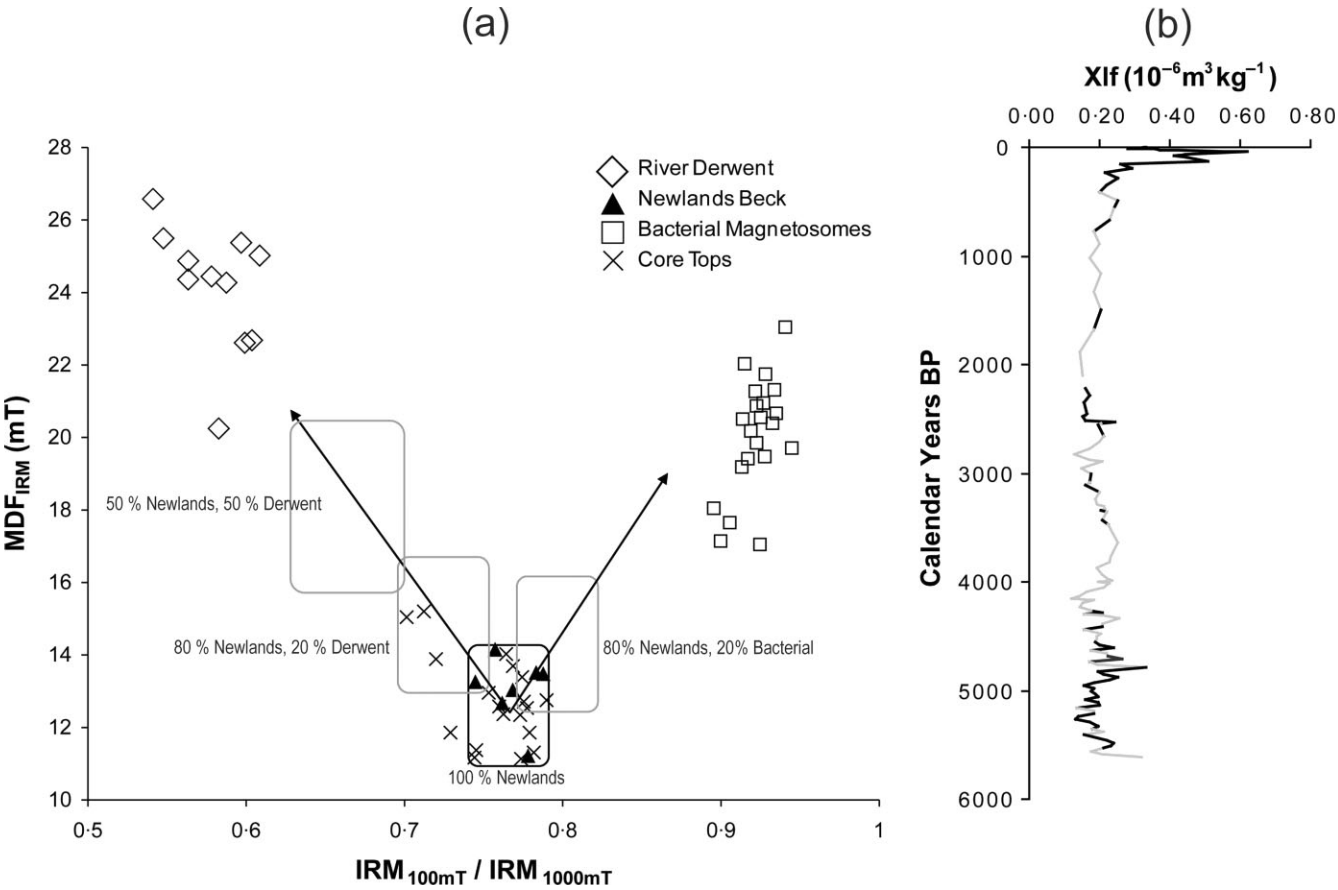

Figure 2. Summary of previous magnetic fingerprinting studies in the Bassenthwaite catchment: (a) contemporary matching of lake 'core top' material to the River Derwent and Newlands Beck suspended sediments showing >>80% contribution from the Newlands subcatchment and precluding any authigenic, in-lake magnetic components (adapted from Hatfield and Maher, 2008) and (b) quantitative determination of the flux and source of lake sediment material over the mid-late Holocene using fuzzy clustering. Magnetic susceptibility is used as a proxy for sediment flux and shows unprecedented 3× increases in the last 100 years. Black lines indicate a dominant source through Newlands Beck and grey lines material sourced dominantly through the Dewent (taken from Hatfield and Maher, 2009).

and immobilized in 10 cc plastic pots using plastic film and cotton wool prior to magnetic analyses. Particle size-specific magnetic measurements included low field, initial magnetic susceptibility (χlf), susceptibility of anhysteretic remanence [χ_{ARM}, i.e. the anhysteretic remanent magnetization (ARM) normalized by the direct current (d.c.) bias field] and step-wise acquisition and demagnetization of isothermal remanent magnetization (IRM). The ARM was imparted using a Molspin demagnetizer with ARM attachment at 80 milliTesla (mT), with a 0·08 mT d.c. biasing field superimposed. IRM acquisition was incrementally imparted in d.c. fields of 20, 40, 50, 100, 200 and 300 mT, using a Molspin pulse magnetizer, and at 500, 700 and 1000 mT (regarded as the saturating field), using a Newport electromagnet. The saturation remanence (SIRM) was then stepwise demagnetized in tumbling alternating current (a.c.) fields of 5, 10, 15, 20, 30, 40, 80 and 100 mT. All remanence measurements were made on a Molspin Minispin fluxgate magnetometer [noise level ~5×10^{-5} Am^{-1} (~5×10^{-10} Am^2)] and all data are expressed on a mass-normalized basis. Information on the major magnetic measurements, ratios and terminology is further detailed in Table I. All magnetic measurements were made at the Centre for Environmental Magnetism and Palaeomagnetism (CEMP) at Lancaster University. Statistical matching of soils, suspended sediments and contemporary lake material was performed using the fuzzy clustering program of Minasny and McBratney (2002). Clustering aims to classify discrete but diverse sample properties in multivariate space and requires no *a priori* knowledge about any of the samples (e.g. geographic location), Fuzzy analysis enables the degree of affinity between a sample and all other clusters to be estimated, rather than categorical assignment of samples to one cluster, as in conventional hierarchal cluster analysis. The fuzzy algorithm aims to minimize within-class sum square errors, calculating the overlap between groups as the 'degree of fuzziness', and provides differing outputs over a range of cluster solutions (Minasny and McBratney, 2002). Selection of the optimal number of clusters is aided by the minimization of two clustering statistical indicators, the FPI (Fuzziness Performance Index) which estimates the degree of fuzziness generated by a specified number of classes, and MPE (Modified Partition Entropy) which estimates the degree of disorganization created by a specified number of classes (Minasny and McBratney, 2002). The optimal solution provides cluster statistics including centroids and a quantitative affinity of each sample not only to its own cluster, but all clusters ranging between zero (no affinity) and one (identical). Such an approach is preferable in sediment source tracing when samples may consist of mixtures of source materials. To minimize the possibility of spurious matches, potential cluster properties were first evaluated using the non-parametric Spearman's Rank correlation coefficient to ensure they were not auto-correlated. A second stage involved removal of any outliers in the dataset (defined as >3 standard deviations from the mean) following the method of Hanesch *et al.* (2001). Only properties and samples which satisfied these criteria were made available for clustering.

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Table I. Short summary of environmental magnetic parameters and instrumentation

Magnetic susceptibility, (normalized to sample mass) *Magnetic concentration*	The ratio of magnetization induced in a sample to the intensity of the magnetizing field. Measured within a small a.c. field (~0·1 mT, ~2·5× the magnetic field of the Earth) and is reversible (i.e. no magnetic remanence is induced). Roughly proportional to the concentration of strongly magnetic (e.g. magnetite-like) minerals. Weakly magnetic minerals, like hematite, have much lower susceptibility values; water, organic matter have negative susceptibility. *Instrumentation*: single sample susceptibility meter *Units*: $m^3\ kg^{-1}$
Anhysteretic remanent magnetization, ARM or anhysteretic susceptibility, χARM *Ultrafine magnetite*	If a sample is subjected to a decreasing a.c. field with a small d.c. field superimposed, it acquires an anhysteretic remanence. ARM is sensitive both to the concentration and grain size of ferrimagnetic (magnetite-like) grains, highest for grains close to the lower single domain (SD) boundary and lowest for coarse multidomain (MD) magnetic grains (e.g. >~5 μm in magnetite). If ARM normalized for the d.c. field strength (desirable as different laboratories use different d.c. fields), it is termed an anhysteretic susceptibility. *Instrumentation*: anhysteretic magnetizer (max. a.c. field 100 mT, d.c. field often ~0·08 mT); fluxgate magnetometer. *Units*: ARM, A m^2, χARM $m^3\ kg^{-1}$
Saturation remanence, SIRM *Magnetic concentration*	The highest level of magnetic remanence that can be induced by application of a 'saturating' magnetic field (in many laboratories the highest DC field is 1 T, sufficient to saturate magnetite but not hematite or goethite). SIRM is an indicator of the concentration of magnetic minerals in a sample but also responds (albeit less sensitively than ARM) to magnetic grain-size. *Instrumentation*: pulse magnetizer and/or electromagnet; fluxgate Magnetometer *Units*: A m^2
Remanence ratios, IRM_{nmT}/SIRM % *Degree of magnetic 'softness' or 'hardness' (MD versus SD magnetite; magnetite versus hematite)*	A 'soft' mineral (e.g. coarse MD magnetite) will acquire remanence easily, at low fields (e.g. IRM_{20mT}/SIRM of 90%; a 'hard' mineral (e.g. hematite) will magnetize only at high fields (e.g. IRM_{20mT}/SIRM of <5%, IRM 300 mT/SIRM of 30%)
Demagnetisation ratios, MDF_{IRM}, $SIRM_{nmT}$ a.f. % *Degree of magnetic 'softness' or 'hardness' (MD versus SD magnetite; magnetite versus hematite)*	Following application of a SIRM subsequent demagnetization of a sample (e.g. $SIRM_{-100mT}$) in increasing a.c. fields can determine magnetic 'softness' and magnetic 'hardness'. The Median Destructive Field (MDF) of (S)IRM is the field at which a SIRM is demagnetized to 50% of its original value. These measures can help discriminate between MD magnetite and SD magnetite and or magnetite and hematite. *Instrumentation*: variable field a.c. demagnetizer (max. a.c. field 100 mT); fluxgate magnetometer.

Note: Adapted from Maher *et al.* (2009).

Results

Catchment survey

Comparison between the soil magnetic susceptibility data obtained from the *in situ* SM400, the laboratory-based core scanner (MS2C) and the single sample MS2B sensors (Figure 3) reveals generally good down-profile agreement, at different degrees of resolution. Soil compression during coring is evident (Figure 3a), with up to ~50% shortening, compared to the *in situ* profile. This results in elevated MS2C (laboratory) values compared with the SM400 field values (Figure 3a). Figure 4 shows the spatial variation in pH and the *in situ* magnetic susceptibility for the 'topsoil' (5 cm depth) and subsoils (5–20 cm) for the 64 catchment sites. Topsoils are slightly more acidic than the subsoils, especially throughout the upper area of the Newlands Valley, which (together with the peats around Skiddaw) displays the lowest pH values in the catchment. The *t*-tests indicate that the soils of the Newlands and Coledale areas (mean pH = 5·3; standard deviation = 0·4) have statistically lower pH values ($n = 44$; $p < 0·0001$) than the GGD soils (mean pH = 6·0; standard deviation = 0·5), reflecting different soil forming environments within the subcatchments. For magnetic susceptibility, the highest values occur in the subsoils of those soils formed on acid igneous intrusions (e.g. around St Johns in the Vale), reflecting the input of primary magnetic minerals. For the remaining, Skiddaw Slate-dominated areas of the catchment, altitude/soil type appear to be reflected in the pattern of susceptibility, with slightly higher values in the valley and alluvial soils and lower values in the moorland peats and podsols. Thus, the topsoils of the GGD subcatchment, with its higher proportion of lower lying land, display significantly higher ($n = 22$; $p < 0·0001$) magnetic susceptibility values [$0·38 \times 10^{-3}$ SI (±0·28 as one standard deviation)] than much of the steeper and higher Newlands area [$0·13 \times 10^{-3}$ SI (±0·12)].

The 64 down-profile susceptibility records can be split into three main categories (Figure 5): those with relatively uniform and weak ($<0·5 \times 10^{-3}$ SI) profile susceptibility (Figures 5a and 5d, comprising peats, humic and gleyic cambisols and podsols in the Newlands Valley and humic and gleyic cambisols in the GGD); those with relatively uniform and intermediate ($>0·5 \times 10^{-3}$ SI) susceptibility (Figures 5b and 5e, comprising colluvial soils and humic podsols in the Newlands Valley and gleyic cambisols in the GGD); and those with relatively high and varying magnetic susceptibility (Figures 5c and 5f, comprising lower Newlands Valley gleyic cambisols, cambisols and fluvisols in the GGD and cambisols in St John's in the Vale).

The majority of the Newlands profiles (71%) fall into the lowest susceptibility grouping. In contrast, only 23% of GGD soils fall within the lowest group; 58% display intermediate susceptibility values. Notwithstanding slightly differing organic contents, drainage and parent material, many of the GGD soils have near-neutral pH and are broadly classed as cambisols (with various degrees of gleying). Whilst the Newlands Valley

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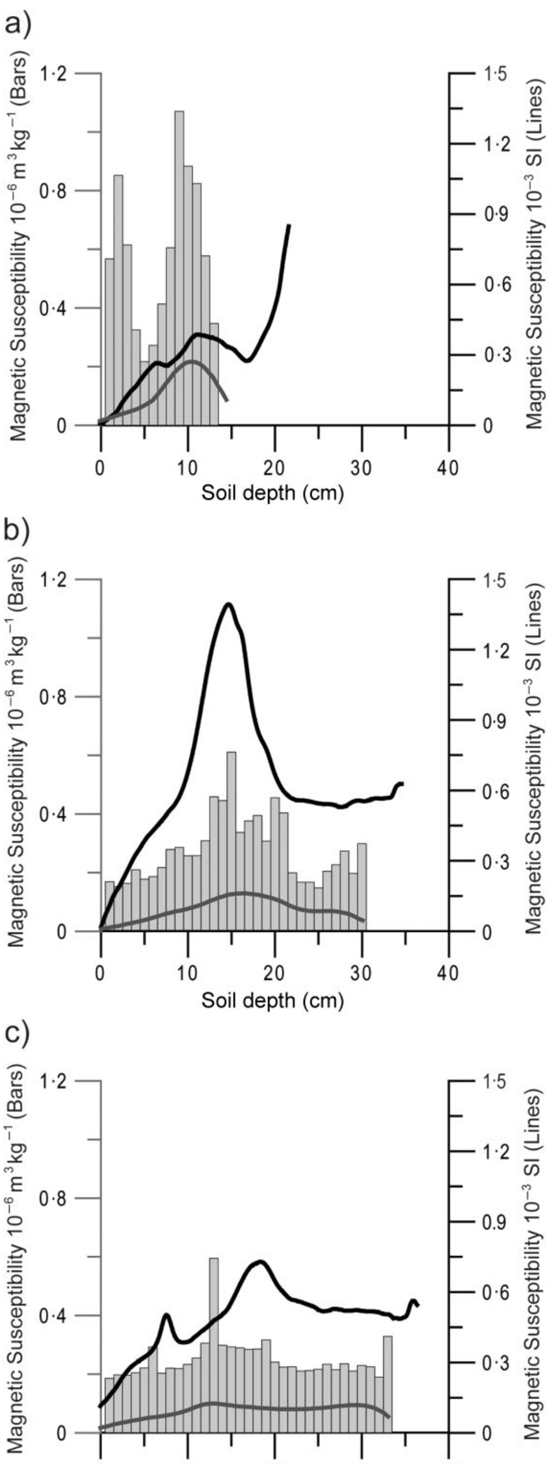

Figure 3. Mass specific magnetic susceptibility (bars) and volumetric magnetic susceptibility measured using the SM400 (black lines) and Bartington MS2C (grey lines) for three samples from (a) the Newlands Valley, (b) GGD subcatchment, and (c) Derwent alluvium.

has some cambisolic profiles located on flatter valley floors and at lower altitudes, there are significant additional areas of humic, histolic and podsolic soils, reflecting the range of drainage and acidity conditions resulting from the varying topography and altitude in this subcatchment. Almost all of the soils throughout the catchment display some degree of intermittent gleying, reflecting the combination of high rainfall and poor drainage. Such conditions are known to restrict soil magnetic enhancement processes and promote dissolution of ferrimagnetic iron oxides like magnetite (Maher, 1984, 1998). In comparison with the range of susceptibility values mapped by Dearing *et al.* (1996) for England and Wales, the majority (84%) of the Bassenthwaite catchment soils (Figure 6) are magnetically weak (<0·4 10^{-6} $m^3 kg^{-1}$), falling within the lowest (but modal) national grouping. With just two exceptions (a humic podsol on igneous rock in the Newlands Valley and a gleyic cambisol under improved pasture), frequency dependent susceptibility (χfd %) averages 3·2% with a standard deviation of 1·5% (national average of 4·1 ± 2·5%, Dearing *et al.*, 1996).

Spanning both the spatial and down-profile magnetic variability in the catchment, 15 soil profiles were selected (Figure 6) for detailed, particle size-specific magnetic analysis.

Particle size variation

Figures 7(a) and 7(b) show the particle size distribution and the relative contribution of each particle size fraction to the concentration-dependent parameters, magnetic susceptibility, ARM and SIRM, for topsoil samples from a Newlands Valley gleyic cambisol and a GGD gleyic cambisol. In the Newlands soil (Figure 7a), clay dominates the particle size distribution (~45%) but it is magnetically the weakest size fraction, resulting in contributions of 30%, 35% and 25% to χlf, ARM and SIRM, respectively. The coarse silt fraction is the strongest magnetically; thus, whilst comprising only 10% of the sample, it contributes 15%, 5% and 17% towards the bulk χlf, ARM and SIRM, respectively. In the GGD soil (Figure 7b), the sand fraction comprises 51% of the sample and contributes 73%, 51% and 62% to the total χlf, ARM and SIRM, respectively. Magnetic grain size and mineralogy also display particle size dependence. Figure 7(c) shows the proportion of SIRM (acquired at 1 T) remaining for each of the five particle size fractions when demagnetized in steps up to 100 mT. None of the soils fully demagnetize in fields of 100 mT, indicating the presence of magnetically 'hard', high-coercivity minerals (e.g. haematite and goethite) in all of the samples. These hard magnetic components comprise 17–77% of the HIRM. The remanence acquired beyond 100 mT but a.c. demagnetized at 100 mT reflects the presence of either fine-grained haematite and/or of maghemite (Maher *et al.*, 2004; Liu *et al.*, 2002). The GGD soil displays the hardest magnetic behaviour. The sand, coarse and medium silt fractions all behave similarly; the median destructive field of their remanence (MDF_{IRM}, i.e. the a.c. field required to remove half the acquired remanence) is 30–32 mT. The fine silt and clay are very 'hard', with MDF_{IRM} values of 62 mT and ~190 mT, respectively. The clay fraction acquires 67% of its SIRM above 300 mT, 40% above 500 mT, suggesting the presence of relatively fine grained (<<2 μm) haematite concentrations (Thompson, 1986; Maher *et al.*, 2004), an inference supported by the 'redness' of this soil (Figure 6b). The Newlands soil is the softest magnetically, with MDF_{IRM} values of 9·5 to 15 mT. Its sand fraction is most resistant to demagnetization; its coarse silt fraction displays the softest behaviour.

Figure 8 summarizes some of these differences in soil magnetic mineralogy on a particle size-specific basis for the coarse silt fraction of each of the 15 soil profiles. The two parameters shown, MDF_{IRM} and $SIRM_{-80mT}$, are sensitive to the degree of magnetic 'hardness' of the samples, determined from their response to incrementally increasing demagnetizing fields (see Table I). The soils show quite variable magnetic behaviour but topsoils are generally softer than subsoils, and soils from the two subcatchments appear to form a number of groupings. Topsoils from the Newlands Valley tend to show the softest,

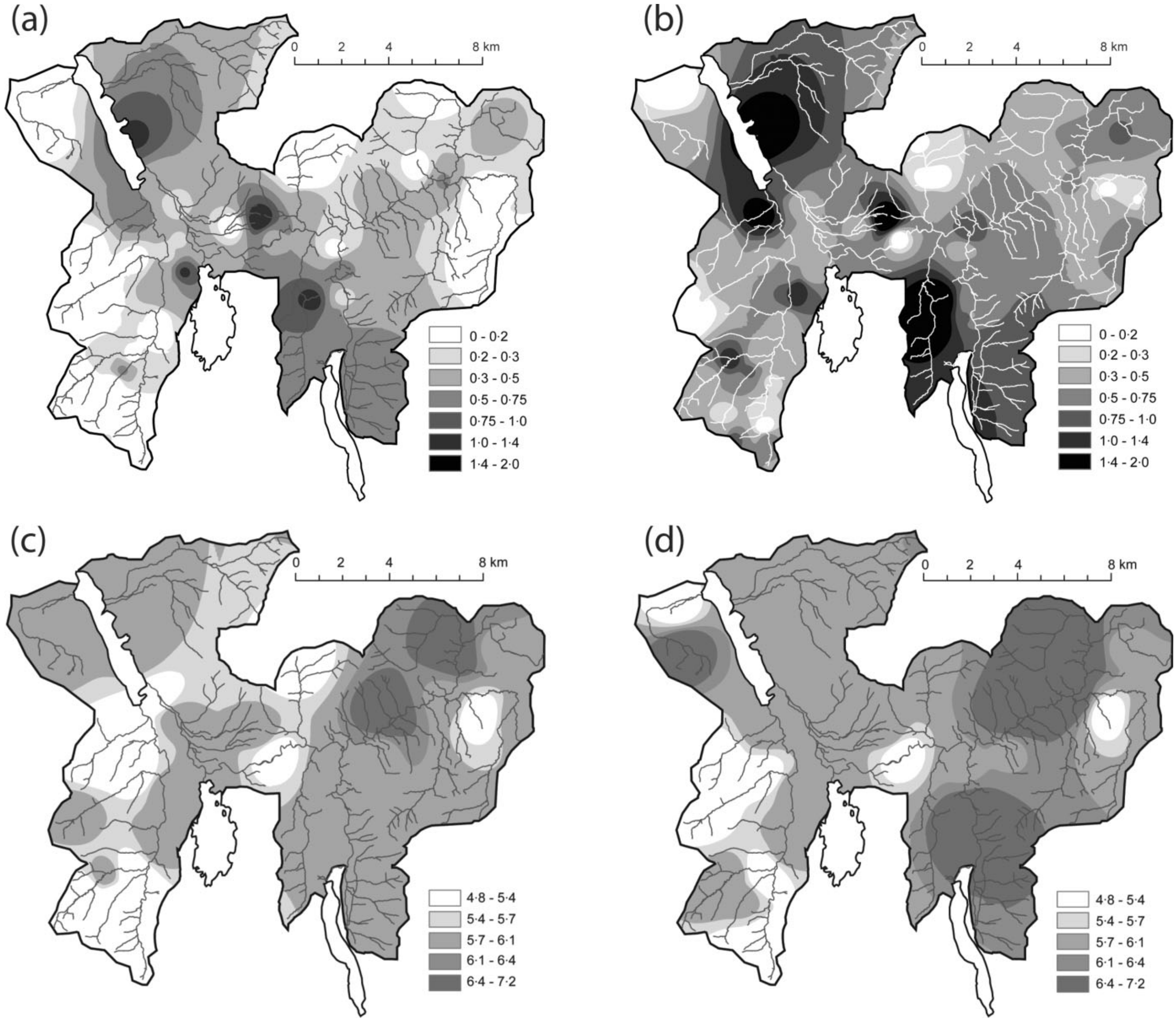

Figure 4. Spatial distribution of topmost (a) and subsoil (b) magnetic susceptibility and topmost (c) and subsoil (d) pH.

most consistent magnetic behaviour (lowest MDF_{IRM} and $SIRM_{-80mT}$ values). Several of the GGD cambisol topsoils (MDF_{IRM} and $SIRM_{-80mT}$ values >10 mT and 10%, respectively) are slightly softer than their subsoil counterparts. These subsoils, together with some Newlands subsoils, form another and harder group ($MDF_{IRM} > 15$ mT). Two further groupings are apparent: two Newlands subsoils and one GGD topsoil (with MDF_{IRM} and $SIRM_{-80mT}$ of ~19 mT and 19%, respectively); and an extremely magnetically hard group, comprising two GGD and one Newlands subsoil samples (MDF_{IRM} and $SIRM_{-80mT}$ values of >25 mT and 25%, respectively). Unsurprisingly, for these coarse silt fractions, soil type seems to play a relatively minor role in determining the magnetic mineralogy; rather, parent material variations within the catchment appear to be a key magnetic determinant.

Sediment–source matching

The magnetic properties of different soils across the Bassenthwaite catchment vary both within and between subcatchments. The soil magnetic properties are also particle size-dependant, as previously reported for the suspended sediments (Hatfield and Maher, 2008). Hence, magnetic tracing of sediments and potential soil sources is likely to be most effective if performed on a particle size-specific basis. Many of the Bassenthwaite soils show little evidence of soil magnetic enhancement, such that, as shown in Figure 7, the coarser (silt and sand) fractions contribute significant proportions of the magnetic signal. These size fractions can thus be used for magnetic 'fingerprinting' of different potential soil source areas and resultant suspended and lake sediments. These fractions are also least affected by overprinting by in-lake, post-depositional processes of magnetic mineral authigenesis, e.g. by bacterial magnetite formation (Hatfield and Maher, 2008). Thus, direct comparison of the coarse silt (31–63 μm) fraction of the different catchment soils can be made with the coarse silt fractions of the inflow suspended sediments (collected roughly on a monthly basis over a three-year period; Hatfield and Maher, 2008) and with the lake sediments contained within six one-metre cores and two three-metre cores (providing a 6000 year record of sedimentation; Hatfield *et al.*, 2008, Hatfield and Maher, 2009), in order to perform quantitative source unmixing. Here, to make most effective discriminatory use of the multidimensional

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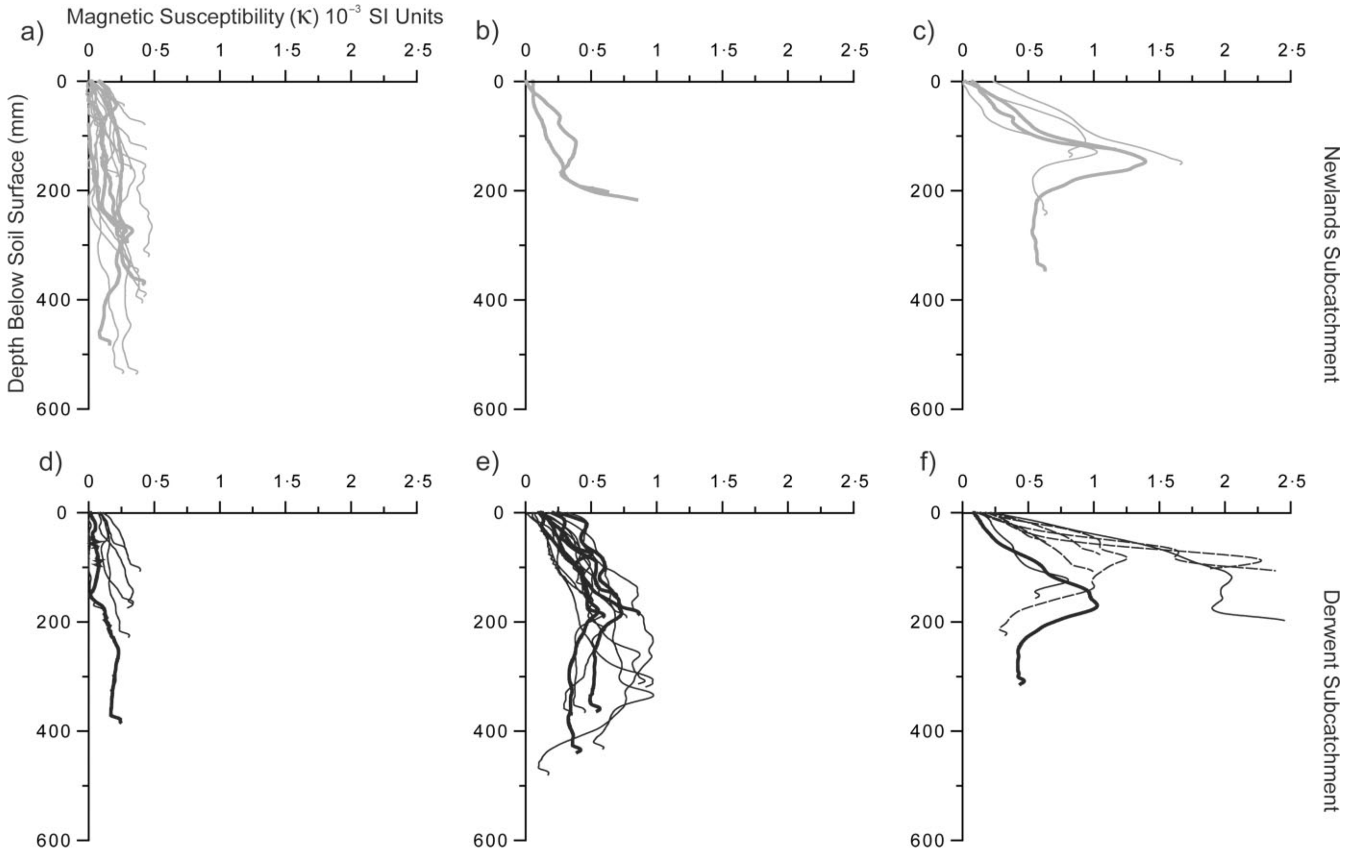

Figure 5. SM400 magnetic susceptibility profiles for the Newlands (grey lines) and GGD (black lines) subcatchments, classified according to susceptibility range; $<0{\cdot}5 \times 10^{-3}$ SI (a and d), $0{\cdot}5$–$1{\cdot}0 \times 10^{-3}$ SI (b and e) and $>1{\cdot}0 \times 10^{-3}$ SI (c and f). Heavier lines indicate the subset of 15 profiles selected for further detailed measurement.

magnetic data, fuzzy-cluster analysis was applied to the multi-parameter magnetic datasets, in order to identify any sample groupings and affinities between the catchment soils, the suspended sediments and the lake sediments. For all of these samples, three magnetic properties of the coarse silt fraction were used (MDF_{IRM}, $SIRM_{-80mTa.f.}$ % and IRM_{20mT}/IRM_{1000mT}, see Table I), in order to match samples on a particle size-specific basis. So-called 'fuzzy' clustering aims to classify samples in multivariate space into clusters, but also provides an affinity of each sample to the cluster groupings, rather than forcing a sample into just one cluster (Vriend *et al.*, 1988). Minimization of the clustering performance indicators (FPI and the MPE) for the soils and for the whole sample set (soils, suspended sediments and lake sediments) suggests the optimal solution consists of six clusters. Figure 9(a) shows the cluster memberships of the 15 soil profiles (topsoils and subsoils); Figure 9(b) the cluster membership of the soils, suspended sediments and lake sediments. Table II shows the cluster centroid values and a description of the cluster magnetic characteristics; also shown are the soil samples dominantly affiliated to each cluster (and their soil type). To validate this clustering solution, Table III shows *t*-test results comparing the MDF_{IRM} and $SIRM_{-80mTa.f.}$ properties of each potential source cluster with the other potential source clusters. All but six of the 42 *t*-tests display significant differences between the source clusters (at the 0·05 level of confidence, 95%); 30 are significant at greater than the 0·01 (99%) level of confidence. Those few clusters not displaying significant differences for MDF_{IRM} do show significant variance for $SIRM_{-80mT\ a.f.}$ (and vice versa) and generally form minor clusters (1, 2 and 3) with low sample affiliations (Figure 9b). The majority of the sediments show affinity with Clusters 4, 5 and 6 (Figure 9b), which are statistically very different from each other, or, in the case of the GGD subsoils, form one unclassified sample group.

As shown in Figures 9(a) and 9(b), the affinity of individual samples to their ascribed cluster is generally very strong; few samples show affinity to more than one cluster. Cluster 6 mainly contains topsoil samples from the upper and middle Newlands Valley, together with a subsoil sample from Keskadale, two GGD topsoils from Glenderaterra and St Johns in the Vale and a soil from the Greta Valley. Cluster 4 showing slightly harder magnetic behaviour, mainly contains the subsoils of the Newlands and two GGD subsoils. Cluster 5 comprises a single soil, a GGD (Glenderamackin) subsoil. Clusters 1, 2 and 3 group several of the harder upper and lower Newlands Valley and Coledale subsoil samples with several of the Glenderamackin and Troutbeck GGD topsoil samples. The extremely hard subsoil samples in the upper Newlands Valley, Glenderaterra and Troutbeck form a disparate, unclassified group.

Figure 9(b) shows the results of fuzzy cluster analysis of all the samples; the soils, inflow suspended sediments and Bassenthwaite Lake sediments (core-tops and down-core). The majority of the Newlands suspended sediments and the lake sediments samples (including the core-top and recent samples) show high affinity (>90%) with Clusters 6 and 4. A small proportion (16%) of the GGD suspended sediments, and the older lake sediments, are strongly affiliated to Cluster 5, the Glenderamackin subsoil cluster (Figure 9a), and an even smaller number to Cluster 6. The remainder and majority of the GGD suspended sediments show no affinity to any of the clusters, forming instead a discrete, unclassified group.

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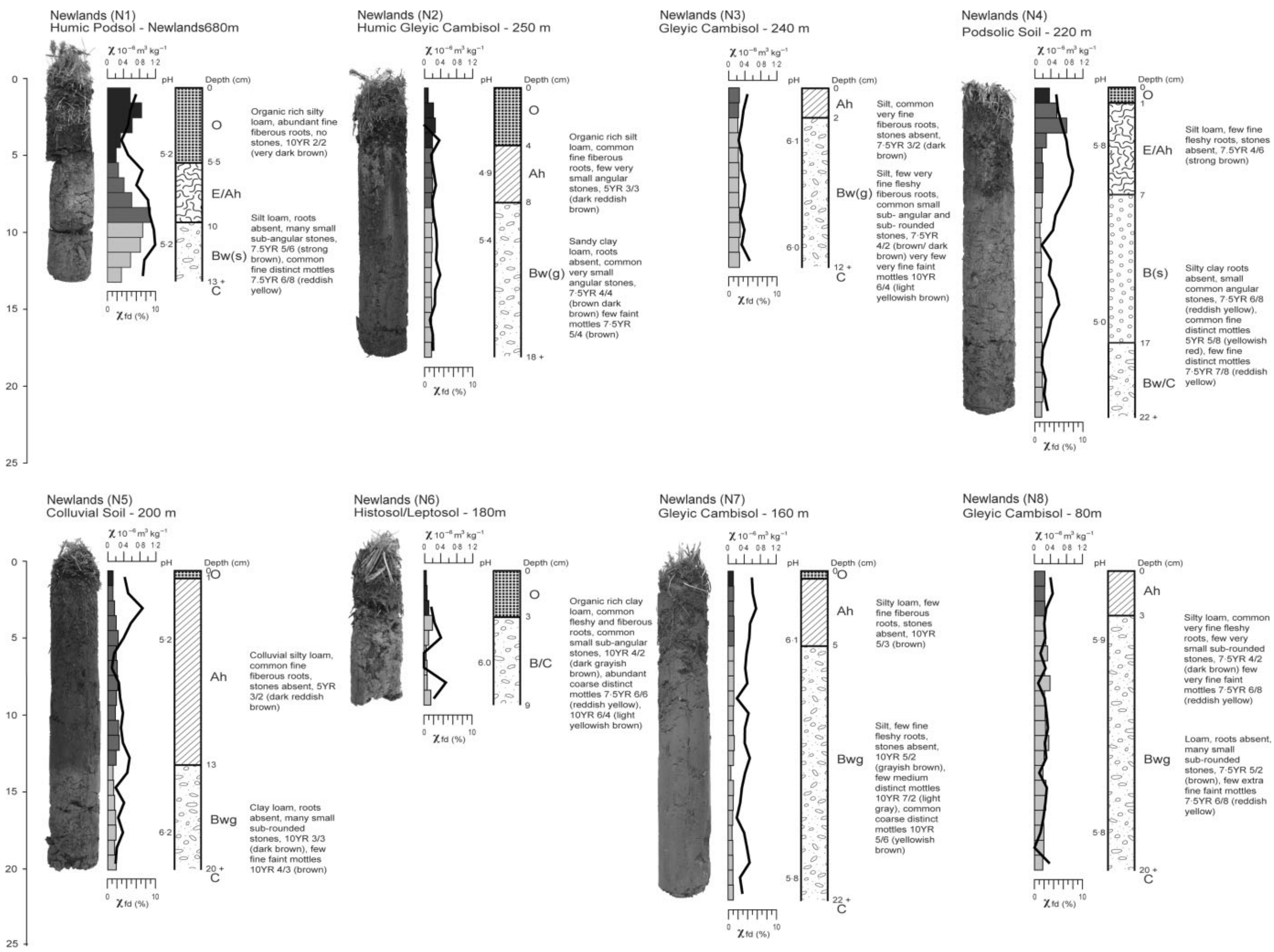

Figure 6. (a) Cores, mass specific magnetic susceptibility, frequency dependent magnetic susceptibility, soil profile description and pH measurements for the seven selected representative profiles from the Newlands sub-catchment. Cores are ordered by altitude.

Table II. Cluster centroids, mean magnetic susceptibility and magnetic and soil characteristics for the five soil and sediment clusters

Cluster	IRM_{20mT}/IRM_{1000mT}	MDF_{IRM}	$SIRM_{-80mT}$	Soil sample identifiers (see Figure 5)	Soil type
1	0·20	17·7	17·0	N4b, N8b GGD2a	Podsolic (Sub), Cambisol (Sub), Cambisol (Top)
2	0·24	11·3	12·8	N5b, N8a	Colluvium (Sub), Cambisol (Top)
3	0·32	10·0	10·4	N3a GGD3b	Cambisol (Top) Cambisol (Sub)
4	0·19	14·3	11·0	N2a, N6b, N7b GGD5b	Cambisol (Top), Histosol/ Leptosol (Sub), Cambisol (Sub) Cambisol (Sub)
5	0·20	17·2	10·8	GGD3b	Cambisol (Sub)
6	0·22	11·1	6·1	N1a, N3b, N4a, N5a, N6a, N7a GGD1a, GGD4ab, GGD5a	Podsol (Top), Cambisol (Sub), Podsolic (Top), Colluvium (Top), Histosol/Leptosol (Top), Cambisol (Top) Cambisol (Top), Cambisol, Cambisol (Top)
Unclassified				N1b, N2b GGD1b, GGD2b	Podsol (Sub), Humic Gleyic Cambisol (Sub) Humic Gleyic Cambisol (Sub), Cambisol (Sub)

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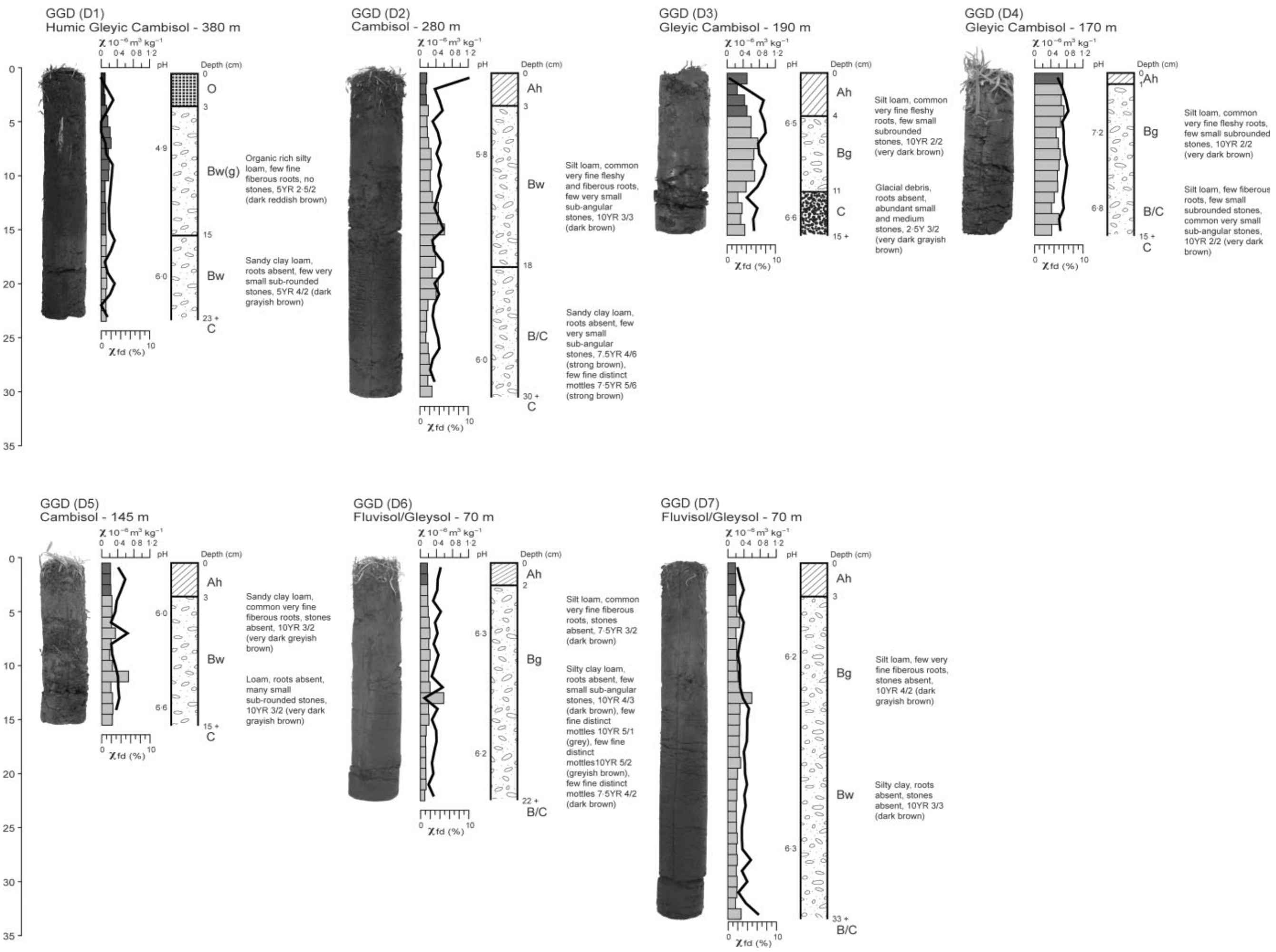

Figure 6. (b) Cores, mass specific magnetic susceptibility, frequency dependent magnetic susceptibility, soil profile description and pH measurements for the eight selected representative profiles from the GGD sub-catchment. Cores are ordered by altitude.

Discussion

The soils within the Lake Bassenthwaite catchment are characterized by rather low magnetic mineral concentrations, with the exceptions of those soils based on localized igneous intrusions. Parent materials are otherwise dominated by the mudstones and siltstones of the Skiddaw Slate Group. With the possible exception of some of the GGD cambisols, which show slightly higher magnetic content and slightly softer magnetic behaviour in their topsoils compared with their subsoils, there is little evidence of soil magnetic enhancement. In addition to the generally rather low magnetic susceptibilities, low values of frequency dependent susceptibility indicate negligible concentrations of the ultrafine ferrimagnetic grains typical of *in situ* pedogenic magnetite formation (e.g. Maher, 1988). Magnetic enhancement processes are likely to be restricted at lower altitudes by the wet climate and slowly draining soils, and at higher altitudes, by excess acidity in the more freely draining podsols (Maher, 1998). Soil magnetic properties appear primarily dominated by detrital inputs and especially by the subtle geological variation between the two major subcatchments, the GGD and Newlands Valley. Weathering of the friable, metamorphosed rocks of the Buttermere Formation of the Newlands Valley has produced soils which are consistently more acidic, and magnetically softer and weaker than those developed on the Kirkstile Formation within the GGD subcatchment.

These factors appear key in affording discrimination between the two subcatchments; Newlands Beck which efficiently transports material through its 'flashy' engineered catchment, and the GGD which appears to store much of its sediment on its floodplain and/or the presently aggrading delta (Hatfield *et al.*, 2008; Hatfield and Maher, 2009). Statistical examination of the affinities between the soils (as potential sediment sources), the contemporary suspended sediments of the different inflows and the lake sediment record enables examination of linkages between them and identification of key sediment source areas within the Bassenthwaite catchment. Critically, strong affinity is evident between the topsoils of the upper and middle Newlands valley and subsoils of the Keskadale area of the Newlands Valley, the Newlands Beck suspended sediments and the core-top and recent sediments of the Lake Bassenthwaite sediments. Independent elemental analysis of the lake and suspended sediments supports these magnetically-derived affinities and linkages (Hatfield and Maher, 2008). Substantial river bank erosion is evident in the Keskadale area at the present day (Figure 10a).

The clustering shows that some soils from the Glenderaterra and St John's Beck areas of the GGD subcatchment (i.e. underlain by localized igneous intrusions) also show affinity with these Newlands soils and sediments. However, they show no affinity with the suspended sediments of the Derwent, indicating that they do not contribute significantly to its contemporary sediment load. In similar vein, suspended sediments from the

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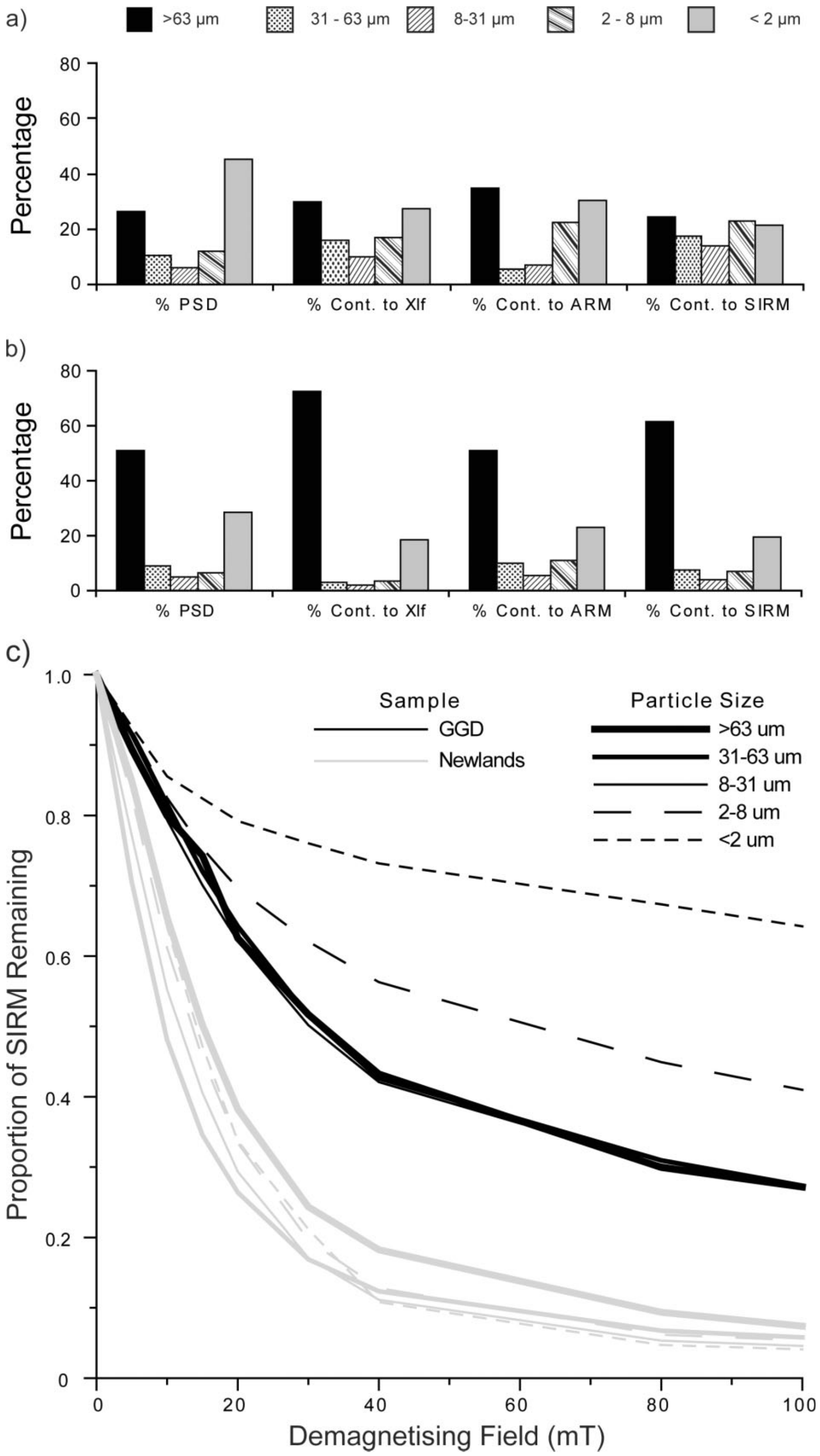

Figure 7. Particle size distribution and percentage contribution to bulk χlf, ARM and SIRM (a, b) and SIRM demagnetization curves (c) for different particle size fractions for topmost material from the Newlands and GGD samples shown in Figure 2.

Mosedale Beck area of the GGD are magnetically too weak, coarse and hard (Hatfield and Maher, 2008) to match with the Derwent suspended sediments. These locally magnetically distinctive 'fingerprints' appear to be quickly mixed with and 'buffered' by the larger sediment loads transported from upstream of the tributary junctions.

None of the suspended or lake sediments shows any affinity with Clusters 1 or 2 (representing the lower altitude cambisols of the middle and lower Newlands Beck area). One lake sediment sample (dated to the medieval period) shows affinity with Cluster 3, which otherwise comprises one soil from the Coledale Beck area.

Table III. The *t*-test confidence levels showing the difference between clusters for two magnetic parameters used in the clustering solution

		MDFIRM (mT)							
		1	2	3	4	5	6	U	
$SIRM_{-80mTa.f.}$ (%)	1		99%	99%	99%	–	99%	99%	1
	2	99%		–	95%	95%	–	99%	2
	3	99%	95%		95%	95%	–	99%	3
	4	99%	95%	95%		99%	99%	99%	4
	5	99%	–	99%	–		99%	99%	5
	6	99%	99%	99%	99%	99%		99%	6
	U	99%	99%	99%	99%	99%	99%		U
		1	2	3	4	5	6	U	MDFIRM (mT)
		$SIRM_{-80mTa.f.}$ (%)							

Note: Values show the percentage confidence a difference exists – values indicate the difference is not significant at the 95% confidence level. For cluster colours see Table II.

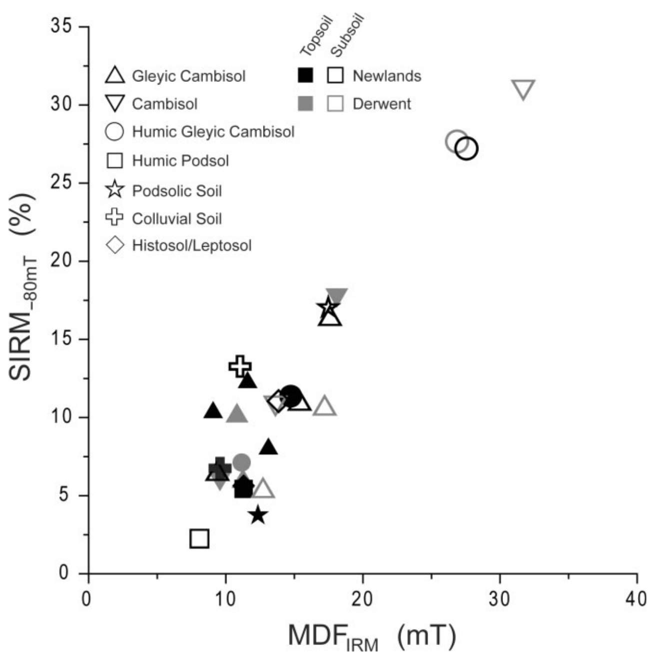

Figure 8. MDF_{IRM} (in mT) versus $SIRM_{-80mTa.f.}$ (in percentages) bi-plots for the 15 topsoils (solid symbols) and subsoil (open symbols) from the Newlands (black symbols) and GGD (grey symbols) subcatchments.

A number of the older Bassenthwaite Lake sediments, namely those from earlier periods of sediment deposition (i.e. pre-nineteenth century), show affinity with a restricted number of the Derwent suspended sediments and a GGD subsoil from the Glenderamackin subcatchment. This suggests that erosion and transport of subsoils from this area provided a significant source of sediment to the deep basin of Lake Bassenthwaite in the pre-nineteenth century period. The late nineteenth century saw peak mining yields, catchment disturbance and an increased availability of highly mobile 'subsoil type' material in the catchment. Sediment accumulation rate increased in the lake during this time (Hatfield *et al.*, 2008) and the cluster solution shows greater affinity to Newlands subsoil material suggesting greater mobilization of sediments from these areas with effective delivery to Bassenthwaite Lake.

It is evident from the large group of unclassified samples (Figure 9b) that the majority of the Derwent suspended sediments have as yet no identified source. It is unlikely that post-erosional/depositional processes have affected these (or any of) the samples as magnetic susceptibility measurements on wet and dry samples show no evidence for authigenesis of magnetic sulphide phases, and bacterial magnetosomes are confined to the clay fraction (Hatfield and Maher, 2008). The Derwent suspended samples are magnetically all very similar, indicating consistency of source despite collection over a range of different time periods and discharges. Their presence as a discrete and unclassified cluster shows that the present soil sample set has not encompassed their source. Given the wide-ranging nature of the catchment soil survey, and that the typical GGD cambisols or the Mosedale, Glenderaterra and St John's Beck soils do not contribute to the Derwent suspended sediments (as described earlier), there remain very few possible (and unsampled) sources. By a process of elimination, the steep and relatively inaccessible slopes of the Troutbeck (and mid- to higher altitude areas of the Glenderamackin) subcatchments are the most probable and as yet unaccounted for source for the suspended sediments carried by the Derwent inflow. These areas may contribute the magnetically very hard mineralogy (represented by the outlying group of subsoils, Figure 9a), possibly at least partially admixed with some softer GGD soils, to produce the characteristic magnetic signature of the majority of the Derwent suspended sediments. As shown in Figure 10(b), river bank erosion is occurring along the Glenderamackin at the present day. In addition, mid- to high-altitude areas around Troutbeck have previously been mapped as being of 'high erosion risk' (Orr *et al.*, 2004).

Identification of sediment sources, in contemporary and historical contexts is essential for assessing the nature and scale of any sediment mitigation measures. Adoption of a nested approach to sediment tracing by first identifying the spatial signature of sediment sources (e.g. Hatfield and Maher, 2008; Hatfield *et al.*, 2008) followed by identification of source types is a key approach in large catchments. Here, magnetic fingerprinting using fuzzy clustering has enabled identification of soil forming environments which contribute most to sub-catchment suspended loading and ultimately to Bassenthwaite Lake. Such information is vital not only for deployment of mitigation measures but for the understanding of upland sediment dynamics especially in response to the extreme land-use and climate pressures felt in these regions.

Conclusions

The soils of the Lake Bassenthwaite catchment generally have low magnetic mineral concentrations, and low values of frequency dependent susceptibility, indicating there is little *in situ* pedogenic formation of magnetite. Only the better drained cambisols of the GGD subcatchment show any evidence of slightly higher magnetic content in their topsoils, whilst both some of the GGD and Newlands topsoils show slightly softer magnetic behaviour compared with their subsoils. High rainfall, slow drainage and/or high soil acidity levels mostly militate against soil magnetic enhancement in this region. Hence, differences in soil magnetic mineralogy mainly reflect differences in geology, even within the area of the Skiddaw Slate Group.

On a particle size-specific basis, the soil magnetic properties enable statistical differentiation between soil groupings from different areas of the catchment. Magnetic parameters sensitive to the degree of magnetic hardness prove most discriminatory in this context. In turn, these soil magnetic clusters can be examined in terms of possible affinity with the contemporary suspended sediments from the Bassenthwaite inflows and the lake sediment record. The recent lake sediments (post-nineteenth century) show strong affinity with the Newlands suspended sediments and upper and middle Newlands Valley topsoils and a Keskadale subsoil. The older lake sediments show strong affinity with a restricted number of the Derwent suspended

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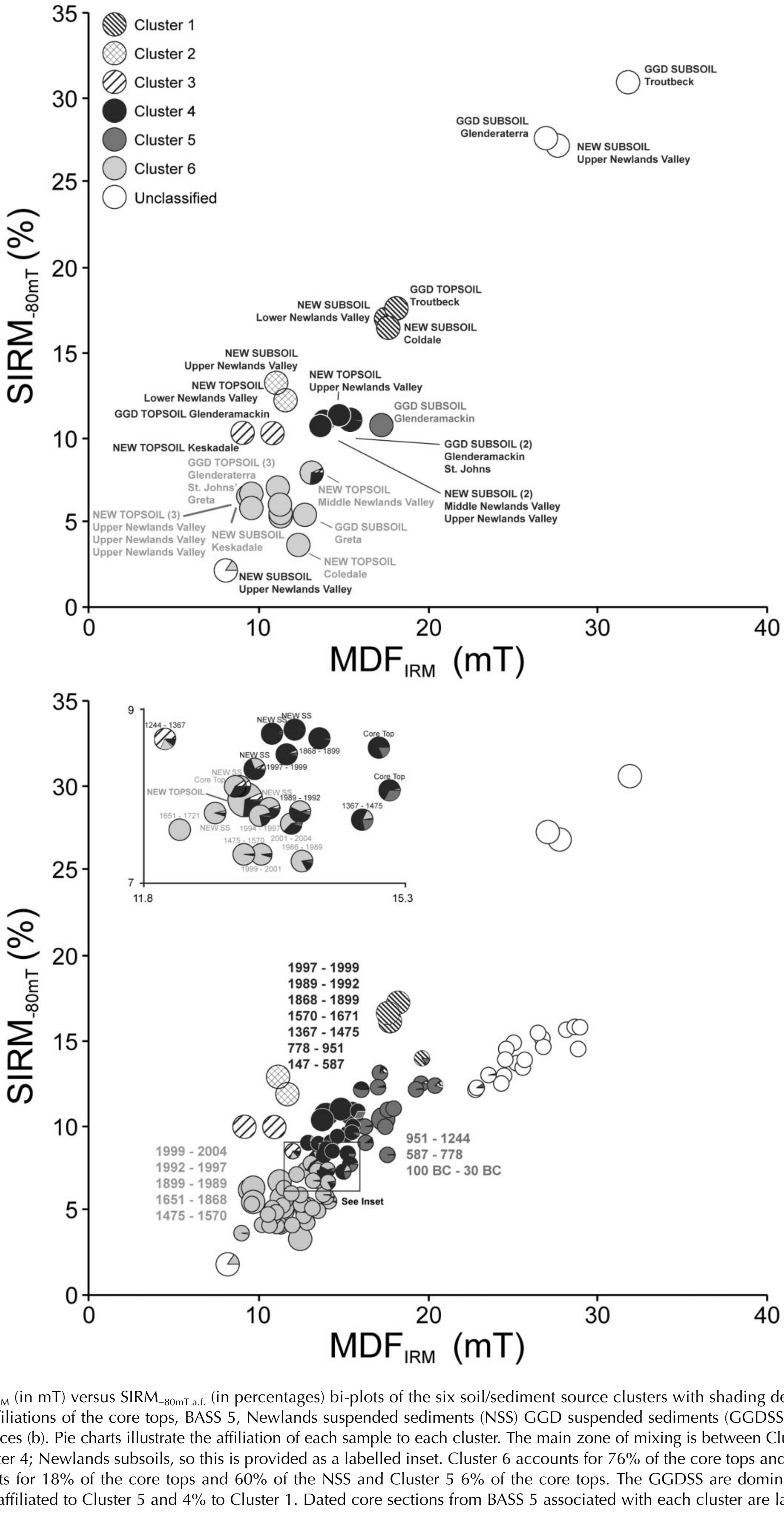

Figure 9. MDF_{IRM} (in mT) versus $SIRM_{-80mT\ a.f.}$ (in percentages) bi-plots of the six soil/sediment source clusters with shading denoting affiliation (a) and cluster affiliations of the core tops, BASS 5, Newlands suspended sediments (NSS) GGD suspended sediments (GGDSS) plotted with the potential soil sources (b). Pie charts illustrate the affiliation of each sample to each cluster. The main zone of mixing is between Cluster 6; Newlands topsoils and Cluster 4; Newlands subsoils, so this is provided as a labelled inset. Cluster 6 accounts for 76% of the core tops and 40% of the NSS, Cluster 4 accounts for 18% of the core tops and 60% of the NSS and Cluster 5 6% of the core tops. The GGDSS are dominantly unclassified (72%) with 24% affiliated to Cluster 5 and 4% to Cluster 1. Dated core sections from BASS 5 associated with each cluster are labelled in 9b.

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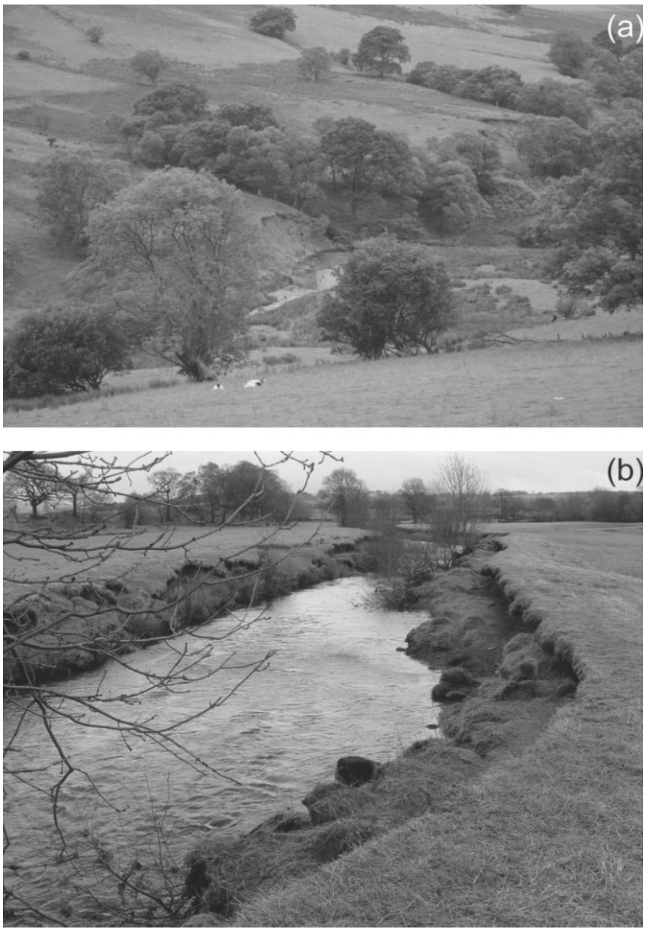

Figure 10. Photographs illustrating the environment and processes: (a) river bank erosion in the Keskadale Valley; (b) river bank erosion along the River Glenderamackin in the GGD.

sediments and a subsoil from the Glenderamackin area. The majority of the contemporary Derwent suspended sediments show no affinity with any of the soil or lake sediment clusters. By a process of elimination, the probable (unsampled) source for these sediments is the mid- to high-altitude areas of the Troutbeck and Glenderamackin tributaries. The Derwent suspended sediments do not appear to contribute to deposition in the deep basin of Lake Bassenthwaite at the present day. They may instead be deposited on the shallow lake delta, on the Derwent flood plain and/or in the eastern shallower segment of the lake basin.

A number of the Newlands suspended sediments show more affinity with Newlands subsoil materials, compared with the recent lake sediments which show stronger affinity with Newlands topsoils. This may indicate enhanced subsurface incision, a process in evidence along the eroding river banks of Keskadale Beck, for example (Figure 10a). This increase in subsoil supply, coinciding with increased rates of lake sediment flux despite recent restrictions on agricultural practices since the 1980s (Orr *et al.*, 2004), may be causally associated with increasing winter rainfall in this region (Malby *et al.*, 2007).

Some of the distinctive, statistically unclassified Derwent suspended sediments may derive from mixing of extremely magnetically hard subsoils from the Troutbeck area with slightly softer soils from lower GGD areas.

This research, building on the work of Hatfield and Maher (2008, 2009) and Hatfield *et al.* (2008), has important implications for science and management of upland and other temperate regions. Magnetic fingerprinting in the Bassenthwaite catchment has been shown to facilitate rapid characterization and matching of sources and sinks, often in environments deemed too homogeneous for geochemical or radionuclide discrimination i.e. within geological units. Such information is critical to catchment managers seeking cost effective targeting of mitigation measures aimed at reducing sediment erosion and delivery and its associated impacts on ecology and geomorphology in marginal and fragile ecosystems.

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