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Application of sediment tracers to discriminate sediment sources following wildfire

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Abstract Wildfire can cause the main sources of fine sediment transported within burned basins to change. Sediment tracing techniques offer an approach to quantify this sediment source response to burning. In this paper, we review the application of various sediment tracers to discriminate sediment sources following wildfire. This includes studies examining changes in fallout radionuclide, geochemical and mineral magnetic properties of burned soil to identify possible fire-related effects on tracer properties and behaviour. Previous work suggests that fallout radionuclides (^{137}Cs, excess ^{210}Pb, ^{7}Be, ^{239,240}Pu) may provide the most effective tracers for post-fire sediment source and budgeting studies. Other tracer properties seem to be overly susceptible to burn effects and might provide a less consistent basis for post-fire source discrimination. The final selection of tracer properties for use in sediment tracing applications should reflect the likely controls of individual tracer concentrations in different sources and their potential response to burning.

Key words wildfire; sediment tracing; sediment sources; fallout radionuclides; river basins

INTRODUCTION

Wildfires represent an important form of disturbance in forest environments that may result in significant changes to runoff and erosion processes, as well as patterns of sediment redistribution in river basins (Shakesby & Doerr, 2006). Fire impacts on erosion and sediment transfer at the basin-scale reflect the balance of sediment inputs from hillslopes and the channel network. Fire-related modifications to soil and surface properties associated with soil heating, loss of vegetative cover and the formation of surface ash deposits can result in substantial increases in hillslope-scale runoff generation and soil erosion (Shakesby & Doerr, 2006). Channel network sediment contributions may also increase in response to elevated post-fire storm flows that are highly erosive (Moody & Martin, 2001). Presently, few studies examine the contributions of fine sediment from major sources to the loadings exported both within, and from, burned river basins. However, there is a need for such information to facilitate better understanding of wildfire impacts on sediment transfer at the basin-scale. Furthermore, this information may also provide the basis for more targeted post-fire mitigation strategies designed to reduce sediment and associated contaminant impacts on water quality. The latter is particularly important for water supply source areas affected by wildfires (Smith *et al*., 2011a).

Sediment source tracing offers an approach to determine the relative contributions of sediment from potential sources. Source tracing techniques have become well-established over the past two decades (Walling, 2005) and have been widely applied in agricultural catchments. More recently, there has been increased interest in the use of source tracers in forest landscapes following wildfires, particularly in southeastern Australia and western Canada. In this paper, we review and synthesise the applications of sediment source tracers in burned forest environments to inform potential users of the different tracers available. On this basis, the objective of this paper is to examine the application of various tracer properties and their source discrimination potential in burned forest environments. We focus on tracers that may discriminate both sediment source types (e.g. surface and sub-surface or channel bank sources) and spatially-defined sources (e.g. burned *versus* unburned, or areas burned at different severities). Tracer properties include fallout radionuclides (^{137}Cs, excess ^{210}Pb, ^{7}Be, ^{239,240}Pu), soil geochemical properties, mineral magnetics and organic compounds. Fire effects on soils may cause modifications to these properties. Therefore, we consider possible fire-related changes to sediment tracers which may affect their source discrimination potential.

FALLOUT RADIONUCLIDES

Fallout radionuclides have been the most frequently used tracers in post-wildfire sediment source and budgeting studies. The environmental occurrence of those fallout radionuclides used as sediment tracers has been extensively described in previous studies (e.g. Walling & Woodward, 1992; Wallbrink & Murray, 1993; Everett *et al*., 2008). In brief, ^{137}Cs (half-life 30.2 years) was produced by atmospheric nuclear weapons testing during the 1950s–1960s. Likewise, ^{239}Pu (half-life 24 110 years) and ^{240}Pu (half-life 6561 years) were dispersed globally following nuclear weapons testing. Excess ^{210}Pb ($^{210}Pb_{ex}$) refers to the amount of ^{210}Pb (half-life 22.3 years) derived from natural atmospheric fallout which exceeds that supported directly by decay of its lithogenic parent ^{226}Ra *in situ*. ^{7}Be (half-life 53.3 days) is another naturally occurring radionuclide that is produced by cosmic ray spallation of nitrogen and oxygen in the upper atmosphere. The fallout of each radionuclide is predominantly associated with precipitation. These radionuclides are typically effective sediment source tracers because of their tendency to strongly adsorb to fine particles, pronounced variation in activity concentration with soil depth, and generally conservative behaviour during transport (Wallbrink & Murray, 1993; Wallbrink *et al*., 1998; Everett *et al*., 2008). However, some fallout may be lost in surface runoff (Dalgleish & Foster, 1996) which could affect the results of sediment budgeting studies that quantify sediment redistribution based on spatially-defined source zones. Such studies assume fallout is retained in the soil and that radionuclide redistribution occurs only by erosion and deposition. This may lead to erroneous results (Parsons & Foster, 2011) but is less of an issue for source tracing studies because they do not require a reference site to estimate the total fallout inventory. Additionally, preferential radionuclide adsorption to fine particles and selective transport generally produce differences in radionuclide concentrations between source soil and sediment samples that are related to particle size (He & Walling, 1996). This also applies to other tracer properties (Horowitz, 1991). As a result, particle size corrections based on specific surface area ratios or analysis of a very fine fraction (typically <10 μm) have been used to minimise the effect of particle size differences on tracer concentrations (Collins *et al*., 1997; Wallbrink *et al*., 1999).

In forested landscapes, maximum concentrations of ^{7}Be and $^{210}Pb_{ex}$ typically occur in surface litter and at the soil surface, respectively, due to the constant production and delivery of these fallout radionuclides. For ^{137}Cs and ^{239,240}Pu, maximum levels tend to be slightly below the surface due to translocation of these fallout radionuclides over the decades since fallout, which is associated with organic matter decay, geochemical diffusion, bioturbation and elluviation processes (Wallbrink & Murray, 1996; Wallbrink *et al*., 2002; Dong *et al*., 2010). All of the fallout radionuclides exhibit activity depth profiles that tend to decrease exponentially with depth in undisturbed forest environments. These distinct depth profiles enable fallout radionuclides to readily discriminate surface and sub-surface (e.g. channel bank) sources of sediment based on differences in activity concentrations (e.g. Fig. 1(a)). Several studies have employed ^{137}Cs, $^{210}Pb_{ex}$ and ^{239,240}Pu to examine hillslope and channel bank source contributions to sediment transported both within and exported from burned catchments (Wilkinson *et al*., 2009; Smith *et al*., 2011b; 2012; Owens *et al*., 2012).

Burning has been reported to increase concentrations of ^{137}Cs and $^{210}Pb_{ex}$ in the surface layers of forest soils (Paliouris *et al*., 1995; Johansen *et al*., 2003; Wilkinson *et al*., 2009), and a similar trend may be expected for ^{7}Be and ^{239,240}Pu. This could allow source discrimination of burned and unburned surface soils, depending on the magnitude of increase, its persistence and the relative contribution of ash to the change in radionuclide activity concentrations at the soil surface. The latter relates to the pre-fire distribution of radionuclides between surface vegetation, soil organic matter and mineral soil. For example, Johansen *et al*. (2003) reported that average concentrations of ^{137}Cs were 40 times higher in ash deposits (192 Bq kg^{-1}; range 7–570 Bq kg^{-1}) and three times higher in the upper 50 mm of burned soils (44 ± 2 Bq kg^{-1}) compared to pre-fire soils (15 ± 3 Bq kg^{-1}) under Ponderosa pine near Los Alamos, New Mexico. The elevated ^{137}Cs levels in the ash layer were largely attributed to burning of the pine needle duff layer that had remained unburned for the prior 60 years and which provided a store of ^{137}Cs accumulated from fallout. In contrast to the conifer forest environment, the pattern of fallout radionuclide distribution appears to differ for eucalypt forest soils in southeastern Australia. While increased post-fire concentrations of ^{137}Cs and ^{210}Pb$_{ex}$ in surface soils under eucalypt forest have been reported (Wilkinson *et al.*, 2009), ^{137}Cs seems to be less associated with ash in contrast to ^{210}Pb$_{ex}$ and ^{7}Be. This reflects the partitioning of ^{210}Pb$_{ex}$ and ^{7}Be between surface litter and soil, with up to 31% and 95% of total inventories stored in surface litter in eucalypt forests, respectively, compared to 15% for ^{137}Cs (Wallbrink & Murray, 1996; Wallbrink *et al.*, 1997). ^{137}Cs in eucalypt forest soils is predominantly associated with mineral soil due to bioturbation transferring decayed litter into the soil following the cessation of ^{137}Cs fallout (Wallbrink *et al.*, 2005; Blake *et al.*, 2009). As a consequence, in eucalypt forests, ^{210}Pb$_{ex}$ and ^{7}Be tracers may be more representative of ash and surface litter, while ^{137}Cs may better represent mineral and organic soil immediately below the ash layer and any remaining O horizon. The apparent difference in ^{137}Cs levels associated with ash formed from the duff layer in a North American conifer forest compared to surface litter in eucalypt forest may reflect greater ^{137}Cs fallout in the Northern Hemisphere (Hodge *et al.*, 1996) combined with slower rates of litter decay and bioturbation in conifer forests under cooler climatic conditions.

Burning may enhance spatial variability in fallout radionuclide concentrations in surface soils. This reflects spatial variations in fuel load and burn patterns that result in varying levels of ash formation and combustion of soil organic material that affects surface soil radionuclide concentrations. Local redistribution or concentration of ash by wind may also be a factor. There is evidence that ^{210}Pb$_{ex}$ concentrations in burned surface soils exhibit greater spatial variability than ^{137}Cs. Smith *et al.* (2012) reported the coefficient of variation (CV) for ^{210}Pb$_{ex}$ and ^{137}Cs measurements of combined surface soil and ash samples from two burned eucalypt forest catchments as well CVs calculated from data presented by two other post-fire tracer studies undertaken in eucalypt forests (Wilkinson *et al.*, 2009; Smith *et al.*, 2011b). The CV range for ^{210}Pb$_{ex}$ concentrations (0.04–0.39) in burned surface soils was found to generally exceed ^{137}Cs (0.06–0.15). This was attributed to the higher concentration of ^{210}Pb$_{ex}$ in eucalypt surface vegetation and litter (Wallbrink *et al.*, 1997) that was transferred to ash, thereby increasing variability in surface soil ^{210}Pb$_{ex}$ concentrations relative to ^{137}Cs. Owens *et al.* (2012) examined temporal variability in post-fire radionuclide concentrations by sampling surface soils in the first and fourth years after wildfire in conifer forest in British Columbia. ^{137}Cs and ^{210}Pb$_{ex}$ concentrations were not significantly different between the two sampling occasions, although mean concentrations were higher in the first year post-fire. Notably, radionuclide concentrations remained significantly greater ($p < 0.05$) in burned surface soil collected four years after the fire compared to soil collected from a nearby unburned catchment. This suggests that fire may have a persistent effect on radionuclide concentrations in surface soils in some landscapes, with clear implications for source discrimination between burned and unburned areas. Importantly, much of the ^{7}Be in surface material could be delivered after burning, given that fires are generally associated with preceding periods of low rainfall during which the inventory of this short-lived radionuclide would decline to low levels. Therefore, ^{7}Be may be less useful for discriminating burned *versus* unburned areas given that, unlike ^{137}Cs and ^{210}Pb$_{ex}$, most of the inventory may be delivered with rainfall after the fire and would not be subject to burn effects.

The effect of fire on fallout radionuclide concentrations and their distribution between ash and surface soils may assist post-fire sediment tracing applications. For source tracing investigations that seek to apportion contributions from hillslope surface and sub-surface (usually channel bank) sources, the tendency for radionuclide concentration to increase in burned surface soils may improve source discrimination. However, the potential for increased spatial variability in radionuclide concentrations from burned surface soils may also contribute to greater uncertainty in estimates of surface source contributions to post-fire sediment flux. Burning does not appear to affect sub-soil radionuclide concentrations based on comparison of measurements of sub-surface soils (>5 cm) and eroding channel banks from burned and unburned catchments (Owens *et al.*, 2012). The partitioning of radionuclides between ash/surface litter (^{210}Pb$_{ex}$, ^{7}Be) and mineral soil (^{137}Cs) observed in eucalypt forests has been used to develop post-fire sediment budgets, based on each radionuclide, that capture the contrasting erosion response of the ash/surface organic layer *versus*

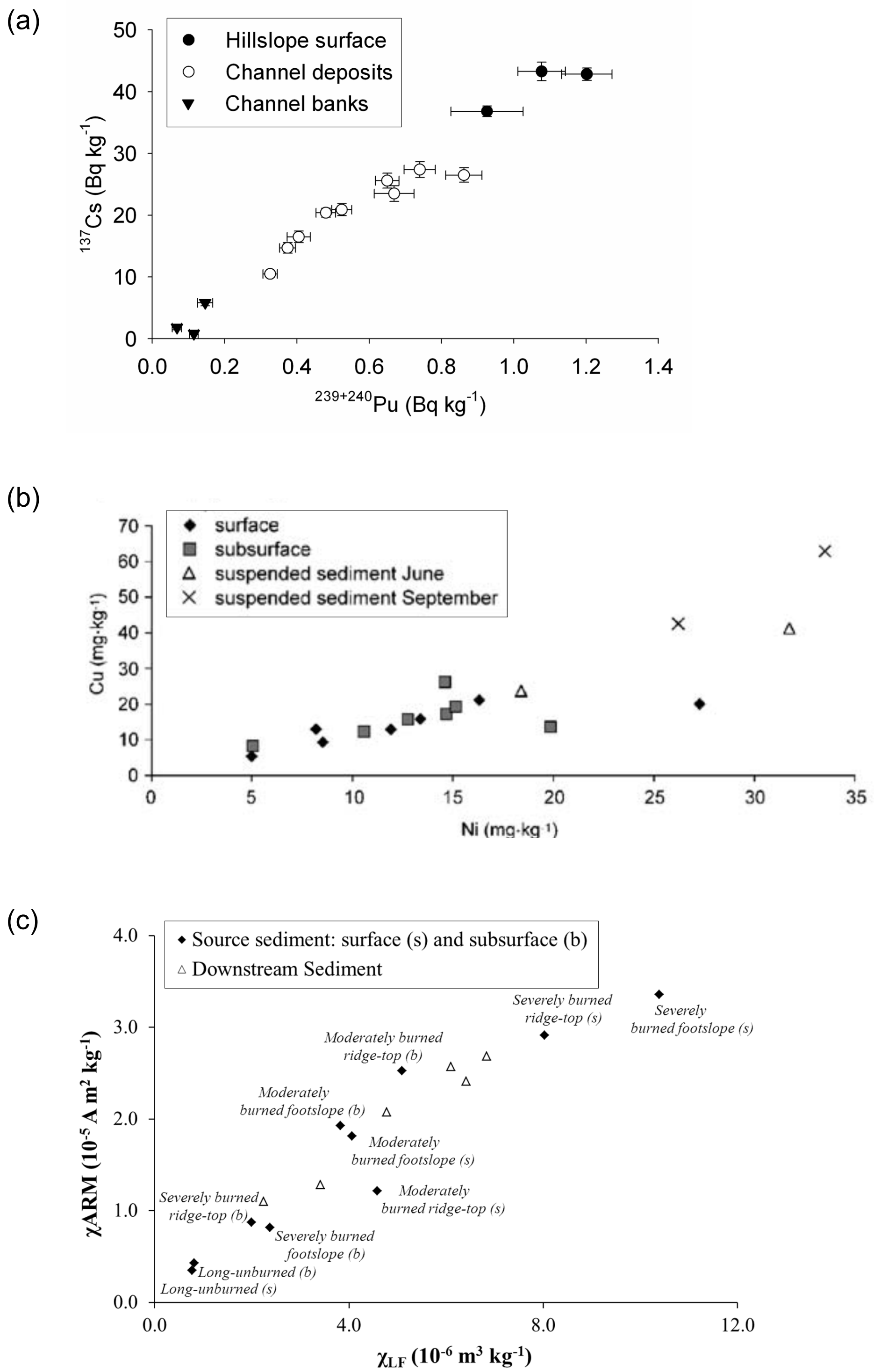

Fig. 1 Examples of bivariate plots comparing various tracer properties from source material with deposited or suspended sediment for: (a) fallout radionuclides ^{137}Cs and $^{239+240}$Pu in composite samples (<10 µm fraction) collected from small catchments affected by post-fire debris flows in northeastern Victoria, Australia (after Smith *et al.*, 2012); (b) geochemical constituents (Cu and Ni) in the <500 µm fraction (with particle size correction between source and sediment samples) from a burned conifer catchment in British Columbia, Canada, in the first year after fire (after Owens *et al.*, 2006); and (c) mineral-magnetic properties associated with soil under eucalypt forest burned at different severities as well as river sediment deposits (all <10 µm fraction) in a catchment near Sydney, Australia (after Blake *et al.*, 2006b).

the underlying mineral soil (Wallbrink *et al*., 2005; Blake *et al*., 2009). The different half-lives and fallout patterns of these radionuclides also provide a basis for examining sediment budgets over timescales ranging from months (immediate post-fire sediment redistribution based on ^{7}Be and ^{210}Pb$_{ex}$) to decades (longer-term patterns of sediment transfer using ^{137}Cs) (Blake *et al.*, 2009).

The presence of fallout radionuclides in ash may have implications for estimating surface source contributions to post-fire catchment sediment exports. The deposited ash layer is transient and may be largely removed within the first year after fire, or sooner, depending on the erosivity of post-fire rainfall and wind events (Reneau *et al*., 2007). Some proportion of ash may also be progressively incorporated into the surface soil through infiltration and mixing associated with bioturbation. As a result, the relative contribution of ash to post-fire catchment exports will change with time since fire. This may present a challenge when characterising surface source radionuclide concentrations based on a single sampling occasion after fire. Changes in the composition (i.e. ash content) of surface material exported from the catchment over time could affect radionuclide concentrations relative to surface material *in situ* comprised of a mixture of ash and soil. The potential significance of this temporal change in surface source signatures will depend on the magnitude of the change relative to spatial variability in tracer properties from the initial sample collection. Given that ^{137}Cs and ^{210}Pb$_{ex}$ concentrations in surface soils have been observed not to be significantly different between the first and fourth years after wildfire (Owens *et al.*, 2012), temporal changes might be of less importance than initial post-fire spatial variability. However, repeated surface source sampling could still be required in some instances and will be essential if ^{7}Be is used in post-fire tracing applications due to its short half-life and ongoing fallout.

GEOCHEMICAL PROPERTIES

Soil geochemical properties have frequently been incorporated in multi-tracer fingerprinting studies of sediment sources, mainly in agricultural or mixed agricultural and urban catchments (e.g. Walling, 2005). The range of potential tracer properties used typically includes various metals (such as Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr, Zn), base cations (Ca, K, Mg, Na) and nutrients (N, P, C) (Walling *et al*., 2008). The selection of a specific geochemical property as part of the final set of tracers used in source ascription is dependent upon establishing statistical differences between target sources for a given study catchment (Collins & Walling, 2002). The potential causes of these differences may relate to natural or human-impacted processes and controlling factors affecting the development of soil profiles. For example, differences in soil geochemical signatures may be influenced by differences in soil parent material, the degree of weathering or by modifications to the soil surface such as by cultivation or burning. Soil nutrients may act as tracers of surface and sub-surface soil based on higher concentrations in surface soils from vegetation decomposition and nutrient cycling.

Soil heating by wildfire can modify surface soil geochemistry (Chambers & Attiwill, 1994; Certini, 2005), thereby resulting in the potential for source discrimination of severely burned, moderately burned and unburned soils (Blake *et al*., 2006a). For example, levels of total Mn in soils have been reported to increase following fire through additions in ash and the physiochemical breakdown of Mn complexed with organic matter (Chambers & Attiwill, 1994), and similar changes might be expected for Fe, Cu, and Zn (Certini, 2005). In contrast, burning has been observed to reduce surface soil total N (Chambers & Attiwill, 1994; Murphy *et al*., 2006), while total P generally does not decrease after wildfire because the volatilization temperature of P (>550°C) is much greater than N (200°C) (Raison, 1979; Hernandez *et al*., 1997; Murphy *et al*., 2006).

The use of soil geochemical properties to distinguish sediment sources in post-fire environments has been explored in eucalypt forest in the Blue Mountains near Sydney, Australia (Blake *et al*., 2006a). In this study, surface soil samples collected from ridge-tops characterized by severe and moderate burn severities as well as unburned conditions were measured for Ca, Mg, P, Pb and Zn using X-ray fluorescence. Blake *et al*. (2006a) found that there was an increase in concentrations of elements in burned relative to unburned soil, with the highest levels reported in the severely burned soil. The increase in concentrations with burn severity was attributed to progressive mineralization of organic matter, such that the organic mass declined in soils burned at higher severities and resulted in increased concentrations of the various constituents measured. These authors concluded that burn effects on soil geochemistry might allow source discrimination of areas of differing burn severity but the “tagged” material largely comprised ash.

Following wildfire in a conifer forest catchment in British Columbia, Owens *et al.* (2006) reported that fire-induced changes to various soil geochemical properties both enhanced and compromised their potential as sediment source tracers. This study focused on the discrimination potential of geochemical tracers between surface (topsoil) and sub-surface (including channel banks) sources (Fig. 1(b)). Various geochemical properties were significantly different between the two sources in both the burned and unburned study catchments. In comparison with the unburned samples, burning resulted in both increases (Co, Cu, K, Ni, P, Zn) and decreases (C, Mn, Pb) in constituents measured in surface material. However, bivariate plots of C *versus* N indicated that a reduction in C concentrations in surface soils, combined with elevated concentrations (exceeding surface soils) in post-fire suspended sediment samples, confounded its use as a source tracer (Owens *et al.*, 2006). The increased C concentrations in suspended sediment may reflect in-stream modifications, such as biofilm development (Petticrew *et al.*, 2006). Similarly, comparison of Cu and Ni between sources and suspended sediment in both burned and unburned catchments (Fig. 1(b)) did not provide the basis for effective discrimination of surface and sub-surface source contributions to suspended sediment. These findings emphasise the need to consider both background and fire-related controls of tracer properties as well as potential in-stream modifications.

MINERAL MAGNETICS

Mineral magnetic properties are often included as part of multi-parameter tracing investigations (e.g. Russell *et al.*, 2001). The most common types of magnetic minerals are haematite, goethite, maghemite and magnetite (Smith, 1999) and, in soil, they originate from both natural and anthropogenic processes (Oldfield, 1991, 1999). Magnetic minerals can occur as discrete fine grains, aggregated concretions or very fine-grained particle coatings (Taylor *et al.*, 1983). From a source tracing perspective, the origin of the magnetic signal is important. Minerals derived from parent material contribute to the “primary” magnetic signature of the soil, which is often overprinted by “secondary” minerals of pedogenic, bacterial, anthropogenic or pyrogenic origin. The sensitivity of secondary magnetic mineral assemblages to short and longer-term environmental change offers potential for sediment source ascription in burned landscapes. The use of mineral magnetic properties as sediment tracers in burned landscapes is based on the production of fine-grained ferrimagnetic minerals in surface soil material during burning (Rummery *et al.*, 1979). Pyrogenic enhancement of soil magnetic properties is linked to changes in oxidation/reduction conditions in surface soil during burning which convert less magnetic iron oxy-hydroxides into more magnetic minerals (Blake *et al.*, 2006c).

The presence of pyrogenic ferrimagnetic minerals, such as magnetite or maghemite, is reflected in enhanced magnetic susceptibility (χ) and saturation isothermal remanent magnetization (SIRM) measurements which are indicative of increased magnetic concentration in burned soil (Oldfield & Crowther, 2007). Mineral magnetic signatures of burned material can be further developed and quantified by looking at magnetic grain-size dependent properties, for example frequency dependent magnetic susceptibility (χ_{fd}) and susceptibility of anhysteritic remanent magnetisation (χ_{ARM}) (Blake *et al.*, 2006b). Quotients of these measures i.e. χ_{ARM}/χ_{lf} or χ_{ARM}/χ_{fd} can be used to distinguish source materials and downstream sediment, and can be usefully compared in bivariate diagrams wherein distinctive envelopes of values due to burning can be identified (Oldfield & Crowther, 2007). Quotients, however, are generally considered to be non-linearly additive (Lees, 1999) which precludes them from use in quantitative source unmixing models.

Recent studies (Blake *et al.* 2006c) have demonstrated spatial variability in mineral magnetic signatures linked to fire severity (Fig. 1(c)). The similarity between the temperature thresholds of magnetic enhancement and changes in soil hydrological properties (e.g. soil water repellency, Doerr *et al.*, 2004) offers potential to link sediment sources related to burn severity to fire impacts on hillslope hydrology (Blake *et al.*, 2006c). Use of concentration-based parameters in this regards shows promise but in many cases source end members represent numerical multiples of each other (Fig. 1(c)), emphasising the need to involve additional properties and/or complementary monitoring or field survey data to allow a more robust discrimination (Blake *et al.*, 2006b).

ORGANIC COMPOUNDS

The use of organic compounds as sediment source tracers is receiving increasing attention with developments in analytical technology. Both the molecular composition of organic compounds and the stable isotope (C and N) signatures of organic matter offer potential in this regard. Sediment tracing applications are in their infancy but recent studies have demonstrated the potential of (i) soil enzyme activities to trace stream sediment to sources under different land use (e.g. Nostrati *et al.*, 2011), (ii) bulk C and N isotopic ratios to explore catchment sediment sources (e.g. Fox & Papanicolaou, 2008), and (iii) Compound Specific Stable Isotope (CSSI) signatures of sediment to explore estuarine sediment sources liked to forestry operations (e.g. Gibbs, 2008) and to link stream sediment to fields of specific crop type (e.g. Blake *et al.*, 2012). Burning of biomass offers potential for further modification of organic compound composition in soil (Atanassova & Doerr, 2011) but the potential for sediment tracing remains largely unexplored. A notable exception in the context of burned soils is a study by Oros *et al.* (2002) which examined the molecular composition of plant and soil organic compounds before and after controlled and wildfire burning. They noted that heating and post-fire leaching and bio-transformations were all important factors in developing organic geochemical fingerprints of burned soils. They concluded that organic geochemical analysis can yield useful chemical fingerprints that identify plant material contributing to organic matter in burned soils and, by extension, this could be a useful soil and sediment tracer. Despite the promise shown by this study, there have been no further studies in this area, perhaps due to the complexity and associated costs of the analyses.

PERSPECTIVE

The application of tracers to investigate sources of fine sediment within burned catchments has considerable potential to contribute to the understanding of post-fire erosion processes and sediment transfer across a range of basin scales and forest environments. However, there are several issues relating to the use of sediment tracers in burned environments that warrant further consideration. These include, amongst others, the need for improved spatial characterisation of tracer properties in source materials. Higher levels of spatial variability in tracer properties from burned forest soils than in other landscape types may necessitate collection of greater numbers of source material samples as well as the use of a composite sampling strategy. In addition, more information is required on tracer partitioning between ash, litter/duff and mineral soil. There is also a need to determine the extent to which tracer properties in burned soil change over time and related to this, repeated post-fire source sampling may be necessary. Lastly, some tracer properties may undergo geochemical alteration as they move between landscape compartments, especially from terrestrial to aquatic systems. Further work investigating the effect of in-stream processes on tracer behaviour and properties is required.

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