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Invited review

Fingerprinting and tracing the sources of soils and sediments: Earth and ocean science, geoarchaeological, forensic, and human health applications

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ABSTRACT

Fine-grained sediment is perhaps the most widespread and pervasive contaminant in aquatic systems reflecting its role in influencing the quality of the water (e.g., turbidity, vector of chemicals and other pollutants) and its detrimental effect on infrastructure (e.g., dams, turbines) and aquatic habitats (e.g., salmonid spawning grounds) through sedimentation. Determining the sources of fine-grained sediment thus represents an important requirement for watershed and coastal management, as well as for understanding the evolution of landscapes and ocean basins. Sediment source fingerprinting utilizes the diagnostic physical, chemical and biological properties (i.e., tracers) of source materials to enable samples of collected sediment to be apportioned to these sources. This review examines the development of the technique within the earth and ocean sciences, focusing mainly on agricultural landscapes. However, the development of new tracers, such as compound-specific stable isotopes, has allowed the technique to be applied in a growing number of environmental settings including forested (including wildfire-impacted forests), urban and estuarine/coastal settings. This review also describes other applications of the fingerprinting approach such as geoarchaeological (e.g., archaeological site formation), forensic (e.g., identifying the sources of soil/sediment particles in criminal investigations) and human health (e.g., identifying the sources of airborne particulate matter, $PM_{2.5}$) applications. Identifying commonalities in methods and approaches between environments and disciplines should foster collaboration and the exchange of ideas. Furthermore, refinement of the sediment source fingerprinting technique requires that several methodological issues be addressed. These methodological issues range from the initial sampling design to the interpretation of the final apportionment results. This review also identifies and assesses these methodological concerns.

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1. Background and history

In recent decades, there has been a rapid growth in the number of studies that have utilized tracing and fingerprinting approaches to investigate the movement of soils and fine sediments in terrestrial and aquatic systems (Koiter et al., 2013a; Walling, 2013; Mabit et al., 2014). This growth is due to the fact that these techniques are able to provide essential information on soil and sediment dynamics that can be used to understand the evolution of landscapes (e.g., Belmont et al., 2007) and assist in river basin management and river restoration (e.g., Owens, 2005, 2008; Evans et al., 2006; Minella et al., 2008, 2014; Walling and Collins, 2008; Gellis and Walling, 2011). In these contexts, the source tracing and fingerprinting techniques have often been part of wider sediment budget investigations (Gellis and Walling, 2011), as the source tracing and fingerprinting techniques alone are sometimes too broad (e.g., topsoil is dominant over channel banks) to enable exact sources (e.g., specific fields, or channel bank reaches) to be determined. Thus, broad classifications of sediment source types can make it difficult to precisely target management strategies intended to control sediment problems. In addition, most sediment source tracing and fingerprinting results are relative (i.e., expressed as percentages), and sediment transport data are often required to convert values into actual sediment fluxes associated with the sources (e.g., Walling and Woodward, 1995; Smith et al., 2011). Source tracing and fingerprinting techniques used in concert with information on sediment transport and sediment budgeting can offer powerful insights into how landscapes behave and can provide important information on geomorphological processes, which, in turn, can be used to guide river basin and coastal management. Mukundan et al. (2012), for example, have demonstrated how sediment source fingerprinting can be used as a management tool for developing total maximum daily loads (TMDLs) of sediment as part of the TMDL programme in the USA.

Early source tracing and fingerprinting studies (e.g., Klages and Hsieh, 1975; Wall and Wilding, 1976) were typically qualitative in nature and concerned with establishing the spatial (e.g., geological) sources of contemporary suspended sediment. These were followed by studies that were more quantitative, again with emphasis on the sources of contemporary sediment (e.g., Peart and Walling, 1986; Walling et al., 1993, 1999; Collins et al., 1997a). Recent developments have seen the technique expanded to include further applications (i.e., new landscape types and research questions, see sections below) and used to determine historical changes in sediment sources using floodplain (e.g., Collins et al., 1997b; Owens and Walling, 2002a; Walling et al., 2003), check dam (e.g., Chen et al., 2016) and lake and reservoir (e.g., Foster and Walling, 1994; Ben Slimane et al., 2013; Pulley et al., 2015) deposits; for a review see D'Haen et al. (2012). The last decade or so has seen an expansion of the types of properties used as tracers and the use of more rigorous statistical approaches and numerical unmixing models.

While there are similarities between approaches concerned with the tracing and fingerprinting of soil and sediment particles in the landscape, there are also some fundamental differences. One useful distinction between the two approaches is that in the case of "tracing" (or "sediment tracing") studies the tracers are pre-selected; in many cases they are applied artificially (e.g., rare-earth elements or fluorescent tracers; Liu et al., 2016). The selection is based on an understanding of the behaviour of that tracer (e.g., fallout radionuclides) and its ability to answer the research questions being investigated. In the case of "source fingerprinting" (or "sediment fingerprinting") studies initially it is unclear what tracers will be selected as fingerprints and samples are analyzed for a range of potential tracers, and statistical methods are used to identify those that are able to discriminate sources.

The term "source tracing" is a hybrid term often used to refer to the use of tracer properties to identify the source of sediments. Thus, the terms "source tracing" and "source fingerprinting" are often used interchangeably to mean the use of the properties of soils and sediments to infer their origins; for simplicity, in this review we mainly use the term sediment source fingerprinting.

While there have been numerous recent reviews of the sediment source fingerprinting approach (e.g., Walling, 2005, 2013; Gellis and Walling, 2011; Mukundan et al., 2012; Guzmán et al., 2013; Haddadchi et al., 2013) most of these are concerned with specific aspects of individual approaches and their application. Thus, Walling (2005) provides an overview of the approach using case study examples from primarily agricultural river basins in the UK. Haddadchi et al. (2013) focus on reviewing sediment tracers and mixing models. The reviews by Gellis and Walling (2011) and Mukundan et al. (2012) are mainly concerned with how sediment source fingerprinting approaches can be used as river basin management and restoration tools, while Guzmán et al. (2013) focus on the provision of information on soil