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Using biomarkers as fingerprint properties to identify sediment sources in a small catchment

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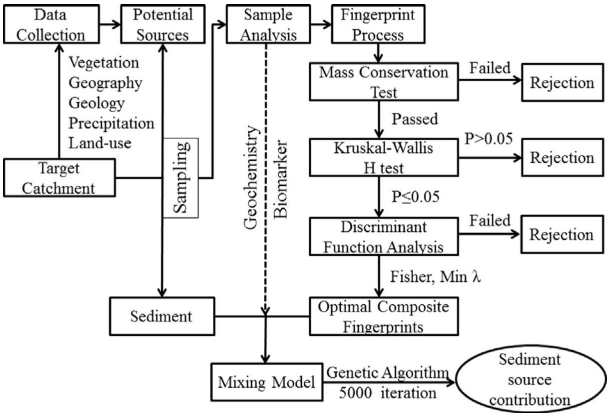
HIGHLIGHTS

- *n*-Alkanes as a new fingerprinting method was used to identify sediment sources.
- Using genetic algorithm finds the best optimum source contribution to sediments.
- Biological and geochemical fingerprinting was used in this study.
- The young forest is the main sediment source.

GRAPHICAL ABSTRACT



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ABSTRACT

Traditional fingerprinting methods are limited in their ability to identify soil erosion sources where geologic variations are small or where different land uses span geological boundaries. In this study, a new biomarker for fingerprinting, specifically, *n*-alkanes, was used in a small catchment to identify sediment sources. The *n*-alkanes were based on land uses, could provide vegetation information, and were relatively resistant to diagenetic modifications and decomposition. This study used a composite fingerprinting method that was based on two types of fingerprint factors (27 biomarker properties and 45 geochemical properties) with 60 source samples (i.e., gully, grassland, forest, and cropland) and nine soil profiles. Genetic algorithm (GA) optimization has been deployed to find the optimal source contribution to sediments. The biomarker results demonstrated that young forest is the main sediment source in this catchment, contributing 50.5%, whereas cropland, grassland and gully contributed 25.6%, 14.4% and 9.5%, respectively; the geochemistry results were similar to the biomarkers. The forest and grassland contributions gradually increased from upstream to downstream, and the sediment contributions of cropland gradually decreased in the direction of the runoff pathway at the check dam. In a comparison of biomarker and geochemical fingerprinting data, the latter may have overestimated the forest inputs to the catchment sediment yields because of a mixed land use history (i.e., forest and grassland). The geochemical fingerprint approach limits its ability to fully discriminate sources based on land management regimes, but the biomarker (individual *n*-alkanes)

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displayed the potential to discriminate between a greater number and different types of sediment sources and to provide greater detail regarding sediment sources.

1. Introduction

Information regarding sediment sources has considerably enhanced our understanding of sediment provenance and the development of catchment sediment budgets (Walling, 2005). Sediment redistribution exerts significant control on the transport and fate of nutrients, organic and inorganic contaminants, and trace or heavy metals (Collins et al., 2010, 2012a; Haddadchi et al., 2013; Zhang et al., 2013). In response to these sediment-related environmental problems, reliable information on sediment sources is critical if mitigation measures are to be targeted effectively. The sediment fingerprinting method has provided a direct and successful approach to quantify sources of sediment from individual river sections to a catchment scale over the previous 30 years (Collins et al., 1997; Walling, 2005; Davis and Fox, 2009; Collins et al., 2010, 2012a, 2012b).

Because of the distinct geophysical and geochemical properties of sediment sources, sediment fingerprinting can help determine the proportion of suspended sediment in a waterway that originates from a specific source within a catchment (Haddadchi et al., 2013; Koiter et al., 2013). Traditional fingerprinting factors have included geochemical factors (Collins et al., 2012a), environmental magnetism (Rotman et al., 2008), rare earth elements (REEs) (Kimoto et al., 2006), radionuclides (Wallbrink et al., 1998), particle shapes (Hatfield and Maher, 2009), and color properties (Martínez-Carreras et al., 2010a, 2010b). However, traditional tracers cannot always distinguish erosion sources by land use when the geologic variations within a study area are small or when different land uses span geological boundaries (Gibbs, 2008; Hancock and Revill, 2013). Current geochemical and geophysical approaches cannot provide vegetation information regarding sediment sources, which represents a major shortcoming of previous research (Blake et al., 2012). Erosion processes are not always related to catchment geology (Hancock and Revill, 2013). Previous studies have indicated that land use is one of the most important factors that directly influence soil erosion (Fang et al., 2012). If new fingerprint properties could improve the relationship between different land use and sediment yield, fingerprinting methods would become more comprehensive.

In particular, *n*-alkane biomarkers provide a potential fingerprinting source. Previous studies have shown that various types of plants produce leaf waxes (*n*-alkanes) with various carbon chain lengths. Long-chain *n*-alkanes (C_{27}–C_{35}) with a strong odd/even predominance are a main component of the epicuticular wax of higher plants (Silva et al., 2012). C_{31} or C_{33} *n*-alkanes dominate in most grasses and herbs, whereas C_{27} or C_{29} *n*-alkanes dominate in most trees and shrubs (Zech et al., 2013). Aquatic algae and microbes are dominated by shorter-chain *n*-alkanes (C_{15}–C_{19}; Meyers (2003)), whereas middle-chain *n*-alkanes (C_{20}–C_{25}) are a dominant component of submerged aquatic macrophytes (Ficken et al., 2000). These plant communities would naturally label the soil and environment where they grow by exuding organic biomarkers because of the leaf waxes and associated *n*-alkanes are not especially water-soluble (Gibbs, 2008; Guzmán et al., 2013). Hydrocarbon molecules are more resistant to diagenetic modifications and decomposition than other forms of organic matter, such as carbohydrates, fatty acids, amino acids and lignin (Matsumoto et al., 2007; Cooper et al., 2015). The former provide long-lived indications of changes in sources of organic matter to a catchment (Meyers, 2003). Based on these theories, the individual *n*-alkanes may discriminate between land uses within a given geologic region.

The loess hilly-gully regions in northwestern China were transported by fierce wind storms during the Quaternary period, and the soil and geologic conditions are generally homogeneous (Tsunekawa et al., 2014). However, the land use/cover in a small catchment usually includes several different landscapes, e.g., forest, grassland, and cropland. In 1999, the Chinese central government initiated a nationwide cropland set-aside program that is known as the Grain-for-Green Project (Fu et al., 2006). The Grain-for-Green Project was developed to increase forest and grassland cover. As a part of this project, vast areas of cropland with a slope gradient that exceeded 25° in mountainous areas were converted to forestland or grassland in the gully and hilly zones of the Loess Plateau. For cropland with other slopes, governments designate a certain quota of cropland in each province every year, and farmers who agree to stop cultivating these lands receive subsidies to cover their loss. Thanks to this income, these farmers will not convert forest or grassland to cropland in the usual manner. The landscape structure has become more stable following the Grain-for-Green Project. Constructing check dams in gullies has been the most widespread and effective strategy to reduce soil and water loss (Shi and Shao, 2000; Wang et al., 2014). Check dams trap all of the sediment that is derived from upstream soil erosion, and the sediment in check dams has a well-documented history of soil erosion. Stable landscapes and sediments that are trapped by check dams provide a source of biomarkers that can be used to identify sediment sources. In the organic geochemistry, *n*-alkanes were the most widely biomarkers, which were used to track the sources of organic matters (Meyers, 2003). However, few studies focused on using the individual *n*-alkanes to identify sediment sources directly, only some researches based on the compound-specific isotope analysis (CSIA) of biomarkers to track sediment sources (Gibbs, 2008; Blake et al., 2012; Hancock and Revill, 2013; Cooper et al., 2015). The latter is often restricted to the instrument, measurement of the individual *n*-alkanes only needs a gas chromatograph fitted with a flame ionization detector (GC-FID), but the CSIA requires to use the isotope ratio mass spectrometer (IRMS) coupled with a GC–Isolink gas chromatograph (GC–IRMS). Guzmán et al. (2013) indicated that the fingerprinting research for the future is the development of tracers requiring inexpensive and rapid analysis approaches that are able to process quickly a large number of samples. It indicated that the individual *n*-alkanes technique was more convenient than CSIA. This paper describes work that was designed to assess the ability of the biomarker fingerprinting technique to discriminate eroded soil sources in a small catchment of the Loess Plateau in northwestern China.

2. Materials and methods

2.1. Study area

The Hujiawan catchment is located in the middle portion of the Loess Plateau between 36.4°N and 36.6°N latitude and between 109.5°E and 109.9°E longitude and covers an area of 27 km^2 (Fig. 1). The elevations within the catchment range from 947 m to 1300 m, and the slope gradient ranges from 0 to 55.3°, with an average of 14.2°; areas where the gradients exceed 10° constitute 75.1% of this catchment (Fig. 2). This region is characterized by a semiarid climate, and the annual precipitation averages 497 mm. Precipitation is mainly concentrated during the rainy season (i.e., from June to September), representing 60–70% of the annual total, most of which occurs in the form of high-intensity rainstorms (Yang et al., 2006). The soil in this region has primarily developed from loess parent materials and has a silty loam texture. The loess layers in the middle Loess Plateau generally have thicknesses of 80–120 m (300–400 m in typical highland areas) and are the thickest known loess deposits in the world, being much thicker than those in Europe and the Americas (Liu, 1985). This