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Biomarkers in sedimentary sequences: Indicators to track sediment sources over decadal timescales

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ABSTRACT

Long-term sedimentary sequence research can reveal how human activities and climate interact to affect catchment vegetation, flooding, soil erosion, and sediment sources. In this study, a biomarker sediment fingerprinting technique based on n-alkanes was used to identify long timescale (decadal) sediment sources in a small agricultural catchment. However, the highly saline carbonate environment and bacterial and algal activities elevated the levels of even-chain n-alkanes in the sediments, leading to an obvious even-over-odd predominance of short and middle components (C_{15}-C_{26}). Therefore, by analyzing three odd, long-chain n-alkanes (C_{27}, C_{29} and C_{31}) in 27 source samples from cropland, gully, and steep slope areas and one sediment sequence (one cultivated horizon and 47 flood couplets), a composite fingerprinting method and genetic algorithm optimization were applied to find the optimal source contributions to sediments. The biomarker fingerprinting results demonstrated that the primary sediment source is gullies, followed by cropland and steep slope areas. The average median source contributions associated with 47 flood couples collected from sediment core samples ranged from 0 ± 0.1% to 91.9 ± 0.4% with an average of 45.0% for gullies, 0 ± 0.4% to 95.6 ± 1.6% with an average of 38.2% for cropland, and 0 ± 2.1% to 60.7 ± 0.4% with an average of 16.8% for steep slopes. However, because farmers were highly motivated to manage the cropland after the 1980s, over half the sediments were derived from cropland in the 1980s. Biomarkers have significant advantages in the identification of sediments derived from different landscape units (e.g., gully and steep slope areas), and n-alkanes have considerable potential in high-resolution research of environmental change based on soil erosion in the hilly Loess Plateau region.

1. Introduction

Sediment sequences document long-term historic records, which could contribute directly to better understanding change in erosional environments in catchment ecosystems (Dearing et al., 2008; Eglinton and Eglinton, 2008; Sritrairat et al., 2012; Zech et al., 2012). These long-term records may provide insight into soil erosion trends (Wang et al., 2014), define reference conditions for soil erosion and sediment delivery models (Zhao et al., 2015), and describe the nature of long-term erosional system dynamics (Franz et al., 2014). However, most existing soil erosion studies have focused solely on inorganic sediment provenance (Yang et al., 2006; Zhang et al., 2006; Collins et al., 2010a, 2010b; Wang et al., 2014; Foucher et al., 2015; Chen et al., 2016b), and the apportionment of organic matter in sediment sequences remains largely unexplored.

Biomarkers such as carbohydrates, fatty acids, alkanols, and lignin are also important carriers of erosional environment information (Rieley et al., 1991; Jia et al., 2008; Bertrand et al., 2013; Fang et al., 2014); however, these biomarkers are limited because of the decomposition of organic matter (Meyers, 2003). For decades, n-alkanes have been well known as valuable biomarkers because of their relative resistance to diagenetic modifications and decomposition (Meyers and Ishiwatari, 1993). The presence of abundant, odd, high-molecular weight ($\geq C_{27}$) n-alkanes indicates mainly terrigenous vascular plant input (Silva et al., 2012), whereas odd, low-molecular weight (such as C_{15}-C_{20}) n-alkanes are characteristic of algae or bacteria (Han and Calvin, 1969; Tu et al., 2000; Meyers, 2003). Odd, mid-molecular weight (C_{21} to C_{25}) n-alkanes tend to originate from freshwater aquatic macrophytes (Ficken et al., 2000; Meyers, 2003). Based on these associations, n-alkanes have proven useful for tracking changes in ecosystems (e.g., autochthonous and allochthonous) at both the regional and global scales (Meyers and Lallier-Vergès, 1999; Fisher et al., 2003; Jeng, 2006; Yunker et al., 2011; Zech et al., 2013; Fang et al., 2014).

Previously, n-alkanes have been used as fingerprint factors to track sediment sources during flood events (Cooper et al., 2015; Chen et al.,

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2016a). In particular, such biomarker fingerprint factors can more comprehensively identify soil erosion sources when geologic variations are small or different land uses span geological boundaries (Gibbs, 2008; Blake et al., 2012; Hancock and Revill, 2013). However, they have been limited as decadal-timescale fingerprint factors used to track sediment sources because when the land use changes, the fingerprint factors in the source soils also change with the plant species. Therefore, in a catchment where land use changes frequently, many portions of the decadal timescale may need to be monitored if using biomarker fingerprinting to track the sediment sources. Research on long-term sediment sources could reveal how human activities and climate interact to affect catchment vegetation, flooding, soil erosion, and sediment sources. Hancock and Revill (2013) (*Hydrol. Process. 27, Page 931*) proposed the following hypothesis: "Biomarkers can be used as fingerprint factors to track sediment sources on long-term (decadal) timescales in areas where the structure of land use is simple and stable." However, no direct evidence has been obtained to confirm this hypothesis.

To control soil and water losses, the Chinese government has constructed a large number of check dams in small agricultural catchments. Currently, >110,000 check dams exist, storing a total of 21 billion m^3 of sediments and 0.0952 Gt of organic carbon on the Loess Plateau (Wang et al., 2011). The main functions of these dams are to stop and store sediments, repair gully beds, raise the eroded base surface, and prevent gully bed undercutting and gully bank expansion (Li, 2001; Xu et al., 2004). The sediments trapped by check dams can serve as natural archives for reconstructing the environmental history of soil erosion in these small agricultural catchments. Furthermore, the sediments trapped behind the check dams differ from natural sediments in two ways. First, they have a fast deposition rate, with tens of meters of sediment typically being stored in check dams within a few decades. Additionally, they have a clear sedimentary sequence, in which the thickness of a couplet varies from a few centimeters to several decimeters (Zhang et al., 2006; Wang et al., 2011; Wang et al., 2014). These characteristics are very rare in natural environments (McConnachie and Petticrew, 2006; Sritrairat et al., 2012; Fang et al., 2014). Check dams play a significant role in carbon and sediment sequestration in the agro-ecosystems of the Loess Plateau (Cao et al., 2009); thus, biomarkers trapped by check dams warrant substantial research attention. These sediments have considerable potential for use in high-resolution research on environmental change, specifically, soil erosion on the hilly Loess Plateau. The main objective of this study is to use n-alkanes as fingerprint factors to reconstruct the soil erosion history and quantify the relative contributions of the various potential sources.

2. Materials and methods

2.1. Study area

The study was conducted in the Nianyangou catchment (37°35′33″ N to 37°35′54″ N, 110°22′4″ E to 110°22′28″ E), which lies in Suede County, Shaanxi Province, in the "coarse sandy hilly area" of the Loess Plateau (McVicar et al., 2005). This agricultural catchment covers a drainage area of 18 ha, and the elevation ranges from 1027 m to 1118 m. The slope gradient ranges from 0 to 35.8%, with an average of 22.3%; areas where the gradient exceeds 25% constitute 66.4% of the drainage area. This catchment is mantled by 5–20 m of Malan loess which is vulnerable to erosion. (Liu, 1985; Hunt et al., 1995). Overall, this catchment is characterized by terrain fragmentation and complex topography (Xue et al., 2011). The concentration of calcium carbonate has ranged from 80.0 g/kg to 162.5 g/kg, with an average of 103.8 g/kg. The climate is temperate continental monsoon, and the mean annual precipitation is 513 mm, of which >70% falls during the rainy season (June to September), typically in the form of high-intensity rainstorms. The frequency of storms has led to severe soil erosion and an extraordinarily high sediment yield in the summer (Xue et al., 2011).

A sediment-trapping reservoir was constructed at the outlet of the Nianyangou catchment in 1960, and it became fully silted with sediments in 1990 (Fig. 1). Additionally, the height of the check dam was repeatedly increased during this period. Cropland covers 77.2% of the study area, with steep slopes accounting for 13.7%, gullies 5.8%, and the check dam only 3.3%. In 1999, the Chinese central government initiated a nationwide cropland set-aside program known as the Grain-for-Green Project (Liu et al., 2012). As a part of this project, vast areas of cultivated land with slope gradients that exceeded 25° (i.e., locations that were most likely to experience severe erosion) were converted to forestland or grassland in the gully and hilly zones of the Loess Plateau (Cao et al., 2009). For cropland areas with other slope characteristics, the government designates a certain quota of cropland in each province every year, and farmers who agree to stop cultivating these lands receive subsidies to cover their losses. Subsequently, cropland areas with steep slopes were converted to fallow land. The topographic map of the study area prior to the construction of the check dam was provided by the Institute of Soil and Water Conservation, Chinese Academy of Sciences and Ministry of Water Resources (Fig. 1, contour lines).

2.2. Sample collection

The land use history was reconstructed from interviews with farmers and a field campaign in 2014. The gullies were very steep, and therefore, the total area of the gullies relative to the total catchment area was small. No fallow land existed in the catchment before 1990, and almost all the land use was cropland except in areas with steep slopes where crops could not be grown by farmers (Fig. 1). Fieldwork involved collecting representative samples of both the source materials and the sediment deposit profiles. In this catchment, the main sediment sources were cropland, gully systems, and steep slopes (i.e., areas with slope gradients $^{>}$20%; Drees et al., 2003), as shown in Fig. 1. The gullies were bare and without vegetation, and the steep slopes were sparsely covered with grass. Source material sampling involved collecting 27 samples of the surface soil from eroding areas representative of gullies (9 sites), steep slopes (9 sites), and cultivated sites (9 sites). For each sampling site, 10 sub-samples were collected at depths of 0–5 cm along transects in a 5 × 5 m grid; these sub-samples were combined in the field to create a single composite sample. These samples were collected using a stainless steel spade, and care was taken to ensure that only material that was susceptible to erosion was collected.

Sediment core samples were collected using an impact drill (HM 1400, Makita, Japan) in the center of the check dam (Fig. 1). The sediment core was carefully sectioned to reveal the flood couplets. The boundary between the couplets associated with individual floods was easily identified because the bottom layer in each couplet was coarse and overlain by a fine layer (Zhang et al., 2006; Wang et al., 2014). Fig. 2 shows an example of several flood couplets collected in the field. Wang et al. (1999) indicated that the concentration of organic carbon in the central section of a check dam was similar to the average concentration of organic carbon in the entire check dam. Therefore, the sediment core was used to represent the average value of the whole check dam. The core sediments were undisturbed, as indicated by the clear water-sediment interface and the preservation of fine sediment laminations. Nearly every flood couplet was composed of one layer of clay and one layer of sand (Figs. 2 and 3); this structure facilitated the identification of each flood couplet. However, not all of the flood couplets exhibited this structure, and some flood couplets had mixed layers (only sand) (Figs. 2 and 3). The thickness of the sediment behind the check dam was measured in situ, and the thickness of a couplet varied from a few centimeters to several decimeters (3–66.5 cm). We collected 48 sediment samples from the sediment core (the first layer was the cultivated horizon) (Fig. 3). Then, we air dried, crushed, and sieved the soil samples through a 2-mm mesh sieve to remove coarse fragments.