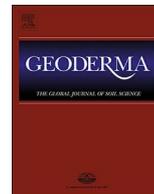




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# Soil carbon redistribution and organo-mineral associations after lateral soil movement and mixing in a first-order forest watershed

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## ABSTRACT

We test the hypothesis that erosion driven soil movement on hillslopes results in an increase in new organo-mineral associations and overall organic matter storage in colluvial deposits within a forested hillslope. We measured mineral specific surface area (SSA), organic carbon (OC), meteoric radioisotopes (<sup>210</sup>Pb, <sup>137</sup>Cs, <sup>10</sup>Be), soil physical properties, C/N,  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$ , and  $\Delta^{14}\text{C}$  in bulk soil and density fractions in a hillslope transect of soil pits. The quantity of OC per unit of mineral surface area (OC/SA) and OC inventories increased by a factor of 2–3 in depositional sites as result of soil mixing due to erosional movement as confirmed by <sup>210</sup>Pb, <sup>137</sup>Cs, and <sup>10</sup>Be profiles and inventories. Soil mixing systematically decreased C/N and enriched stable isotopes of  $\delta^{13}\text{C}$  and  $\delta^{15}\text{N}$ , revealing that formation of organomineral associations instead of microbial processing was responsible for depth trends in organic matter composition. Our findings indicate that the processes that associate organic matter and minerals are fundamentally linked with organic matter composition, and OC/SA, C/N,  $\delta^{13}\text{C}$ , and  $\delta^{15}\text{N}$  provide proxies for organic matter stabilization by soil minerals.

## 1. Introduction

Soil organic matter is the largest active reservoir of carbon on Earth, and understanding the stabilization mechanisms and feedbacks to climate and other anthropogenic change is a critical need to managing and mitigating atmospheric carbon dioxide (CO<sub>2</sub>) (Stocker et al., 2013). Soil carbon can be rapidly transformed into atmospheric CO<sub>2</sub> unless it is protected from microbial oxidation. A key mechanism for retaining organic matter in soils is protection by organomineral associations (Aufdenkampe et al., 2011; Schmidt et al., 2011), in addition to other biotic and abiotic environmental conditions such as freezing (permafrost), burial and anaerobic zones (Schmidt et al., 2011).

Organic carbon (OC) that is relocated within a landscape largely remains in the soil with net gain in carbon storage during transport (Fissore et al., 2017; Harden et al., 1999; McCarty and Ritchie, 2002; Van Oost et al., 2007; Wang et al., 2017). Soil organic carbon buried in depositional settings is often protected by various stabilization mechanisms (Berhe et al., 2007; Doetterl et al., 2016; Kleber et al., 2010; Kramer et al., 2017; Sollins et al., 2006; Van Oost et al., 2007; Wang et al., 2013), and these depositional settings provide thick, moist, fertile soils that increase in situ plant productivity and carbon input to soils

(Yoo, 2005). Interactions between organic matter and minerals—such as occlusion within mineral aggregates or sorption on mineral surfaces (Kleber et al., 2007; Wagai and Mayer, 2007)—shield molecules from degradation (Hsu and Hatcher, 2005; Knicker and Hatcher, 1997) and are more critical than molecular structure for predicting the stability and turnover time of organic matter in soil (Kleber et al., 2007; Schmidt et al., 2011). Confirming mineral association as a protective mechanism, mineral specific surface area (SSA) was found to be a better predictor for OC storage in mineral soils than cation exchange capacity,  $\Delta^{14}\text{C}$ , C/N, clay content, exchangeable oxide minerals, or percent of organic carbon (Berhe et al., 2012).

We use mineral SSA as a proxy for the capacity for a mineral assemblage to protect OC. SSA compiles in a single measurement, the effects of multiple binding mechanisms for organic matter attachment to mineral surfaces and within soil aggregates, including cation bridging, ligand exchange, van der Waals forces, hydrogen bonding, cation and anion exchange, and hydrophobic effects (Keil and Mayer, 2014). For simplicity of discussion we generalize all of these potential interactions as “organomineral associations.”

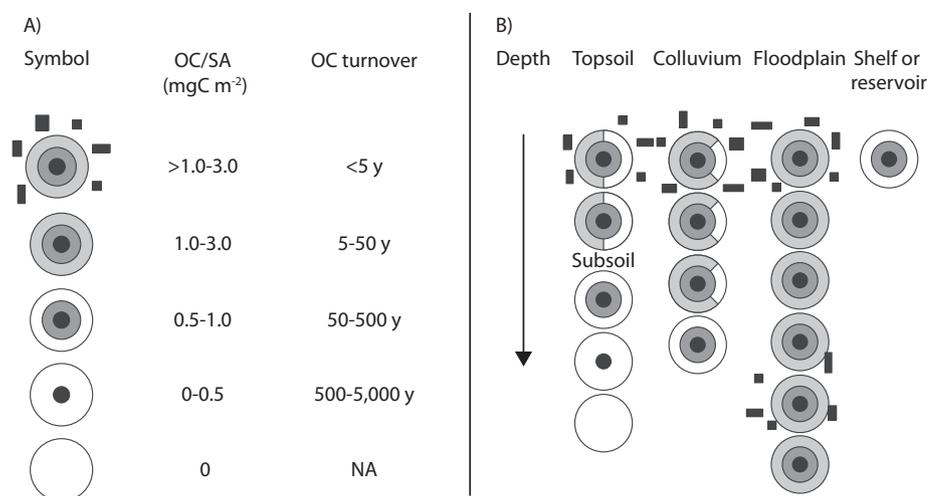
Carbon-mineral associations occur in patches rather than in smooth, homogeneous layers (Mikutta et al., 2009; Solomon et al., 2012;

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Thieme et al., 2010). Organic molecules furthest and most loosely bound to mineral surfaces are observed to turn over more rapidly than organic molecules closest and most tightly bound (Kleber et al., 2007; Sollins et al., 2006; Trumbore et al., 1995), and exhibit characteristic turnover times (Fig. 1A), based on radiocarbon ages from sequential fractionation of soils and oxygen exposure time of marine sediments (Cowie et al., 1995; Hedges et al., 1999; Keil et al., 2004; Kleber et al., 2007; Sollins et al., 2009; Trumbore et al., 1995). We adopt 0.5–1 mg C m<sup>-2</sup> as a range of carbon loading where mineral surfaces provide substantial capacity to protect organic matter, as established by Mayer (1994b), for shallow (< 75 m) ocean shelf sediments over a wide range of locations. Above 1 mg C m<sup>-2</sup>, we expect organic carbon to be less protected by the mineral matrix. Below 0.5 mg C m<sup>-2</sup>, we expect SSA to have additional capacity to protect organic matter and that associated organic carbon is very stable. After Mayer (1994b), subsequent studies corroborated that 0.5–1 mg C m<sup>-2</sup> of carbon loading is a reasonable approximation for organic matter protection by mineral surfaces (Hedges and Keil, 1995; Keil et al., 1994), and established that topsoils can contain organic matter in excess of the ranges observed in ocean shelf sediments (Mayer and Xing, 2001; Wagai et al., 2009).

In seeking to understand the sink or source fate of OC during erosion, three mechanisms were tested: replacement, burial, and respiration (Berhe et al., 2007; Van Oost et al., 2007). We test a fourth mechanism, introduced by Aufdenkampe et al. (2011), that new organomineral associations are created during erosional transport and contribute to carbon sinks. Specifically, we hypothesize (Fig. 1B) that through erosional transport, fresh, reactive organic matter physically mixes with clean, OC-poor minerals from deeper soil horizons to form stable organomineral associations in soils and sediments (Aufdenkampe et al., 2011). Physical mixing has been observed to promote organomineral associations at multiple scales, including hillslopes (Berhe et al., 2012) and soil profiles (Lyttle et al., 2014). We expect that topsoils with excess OC will undergo periods of loss and gain of organic carbon while transported in rivers, deltas, and into the ocean (Hedges and Keil, 1995). In addition to the potential input of reactive minerals in depositional environments, depositional environments often have higher soil moisture and lower pH, which can protonate mineral surface hydroxyls, rendering them less soluble (Brantley and Olsen, 2013) and more attractive to negatively charged organic substrates (Berhe and Kleber, 2013). Repeated burial episodes may also render the organic matter in each depositional sequence less accessible to degradation (Berhe and Kleber, 2013). Not to neglect the significant storage of OC in reservoirs, lakes, floodplains, and wetlands, marine studies of ocean shelf sediments were seminal in examining role of organomineral associations in OC stabilization. Ocean shelf sediments have been subjected to extensive processing in the sediment transport continuum and

**Fig. 1.** (A) Conceptual model of OC protection by organomineral assemblages, where we symbolically summarize published descriptions of organic molecules. The OC furthest and most loosely bound to mineral surfaces (lighter shaded circles) turn over more rapidly than organic molecules closest and most tightly bound to mineral surfaces (darker shaded circles); turnover times are indicated for each range of OC per unit of mineral SSA based on radiocarbon ages from sequential fractionation of soils and on oxygen exposure time of marine sediments (Cowie et al., 1995; Hedges et al., 1999; Keil et al., 2004; Kleber et al., 2007; Sollins et al., 2009; Trumbore et al., 1995). Rectangles represent OC not associated with minerals, or free particulate organic matter. (B) Conceptual model of our hypotheses for depositional profiles, where a key mechanism for organomineral associations is mixing during transport or *in situ*, indicating the depth occurrence of organomineral associations. Symbols indicate ranges of OC/SA and turnover time, where the correspondence with turnover time from part A is less significant when burial in depositional zones protects organic matter, such as on floodplains and ocean shelves.

the OC associated with these sediments represents organic matter that is stably associated with minerals, with published ranges of OC/SA that are consistently 0.5–1 mg C m<sup>-2</sup> (Mayer, 1994a, 1994b; Wagai and Mayer, 2007). This ratio is not as well constrained for soils (Wagai et al., 2009), but floodplain and wetland deposits are known to have very high carbon contents (Noe and Hupp, 2009, Aalto and Aufdenkampe, unpublished data from Peru, Brazil, Bolivia, and Pennsylvania). One missing component among these observations is the hillslope-scale relationship of organomineral associations in colluvium.

In this work we examine SSA, OC, organic matter properties (C/N,  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$ ,  $\Delta^{14}\text{C}$ ) in bulk soil and density fractions, meteoric radioisotopes (<sup>210</sup>Pb, <sup>137</sup>Cs, <sup>10</sup>Be), soil physical properties, and landscape position in a hillslope transect of soil pits. We test our hypothesis that erosion driven soil movement on hillslopes results in an increase in new organomineral associations and overall organic matter storage in colluvial deposits within the landscape. We believe the increase in organomineral associations is the result of a combination of processes, including (1) colluvial redistribution of topsoil with a high concentration of fresh organic matter and of subsoil with a high concentration of clean, carbon-poor mineral surfaces, (2) new formation of organomineral associations due to enhanced physical mixing of organic matter with minerals during erosional transport, and (3) soil chemistry of depositional landscapes altering transported minerals and making them attractive to organic matter. Our work looks at the cumulative effects of all of these processes. We assess on a forested hillslope the distribution of organic matter and mineral specific surface area (OC/SA), along with C/N,  $\delta^{15}\text{N}$ ,  $\delta^{13}\text{C}$ , and radiocarbon to show how erosional transport and mixing processes alter organomineral relationships.

## 2. Methods

### 2.1. Study site

The study site is the Spring Brook watershed (Fig. 2) located in southeastern Pennsylvania, which is the only completely forested watershed with no history of row crop agriculture in the Christina River Basin Critical Zone Observatory (CRB-CZO) (Fisher, 2016; Fisher et al., 2017). Spring Brook is a first order watershed that covers 9.6 ha, with a 250-meter long spring-fed perennial stream. Up gradient from the stream is a 150-meter long, 1-meter deep historic gully that is no longer actively eroding but acts as a depositional swale with a developed A-horizon through the extent of the swale. The soils in Spring Brook are mapped as Manor Series soils (NRCS, 2012). These Typic Dystrudepts are highly micaceous with weak soil structures and coarse loamy texture. Spring Brook is underlain by the Laurels Schist bedrock, a foliated silvery gray-green quartz-plagioclase-muscovite-chlorite-garnet schist

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