



Mobilization of colloidal carbon during iron reduction in basaltic soils



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ARTICLE INFO

Article history:

Received 31 May 2013

Received in revised form 3 January 2014

Accepted 9 January 2014

Available online 7 February 2014

Keywords:

Hawaii

Soil organic matter

Anoxia

Iron reduction

pH

Asymmetric flow field-flow fractionation

ABSTRACT

The transport of organic carbon (C) to deep mineral horizons in soils can lead to long-term C stabilization. In basaltic soils, C associations with short-range-ordered (SRO) minerals often lead to colloid-sized aggregates that can be dispersed and mobilized by changes in soil solution chemistry. In the montane forest region of Hawaii, basaltic soils are exposed to high rainfall and anoxic conditions that facilitate ferric (Fe^{III}) (oxyhydr)oxide reduction. We explored the potential of iron (Fe)-reducing conditions to mobilize C by exposing the surface mineral horizons of three soils from the Island of Hawai'i (aged 0.3, 20, and 350 ky) to 21 days of anoxic incubation in 1:10 soil slurries. Mobilized C was quantified by fractionating the slurries into three particle-size classes (<430 nm, <60 nm, <2.3 nm ≈ 10 kDa). In all three soils, we found Fe reduction (maximum Fe²⁺ (aq) concentration ≈ 17.7 ± 1.9 mmol kg⁻¹ soil) resulted in ~500% and ~700% increase of C in the 2.3–430 nm, and <2.3 nm size fractions, respectively. In addition, Fe reduction increased solution ionic strength by 127 μS cm⁻¹ and generated hydroxyl ions sufficient to increase the slurry pH by one unit. We compared this to C mobilized from the slurries during a 2-h oxidic incubation across a similar range of pH and ionic strength and found smaller amounts of dissolved (<2.3 nm) and colloidal (2.3–430 nm) C were mobilized relative to the Fe reduction treatments (p < 0.05). In particular, C associated with the largest particles (60–430 nm) was dispersed almost exclusively during the Fe reduction experiments, suggesting that it had been bound to Fe-oxide phases. Our experiments suggest that colloidal dispersion during Fe-reducing conditions mobilizes high concentrations of C, which may explain how C migrates to deep mineral horizons in redox dynamic soils.

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1. Introduction

Earth's carbon (C) cycle is influenced by the distribution and behavior of C in soils. Considerable effort is now directed at understanding the mechanisms regulating soil C stabilization and turnover rates (Schmidt et al., 2011) as a function of climate, including temperature, rainfall, and soil saturation (Giardina and Ryan, 2000; Schuur et al., 2001; Townsend et al., 1997; Trumbore, 1997). Many studies observe C deep in the soil profile has longer turnover times than surface soil C (Harrison et al., 2011; Torn et al., 1997). These studies suggest a lower microbial activity and increased association with mineral surfaces—particularly short-range-ordered (SRO) minerals (Chorover et al., 2004; Kramer et al., 2012; Lalonde et al., 2012; Mikutta et al., 2009)—contribute to the persistence of C at depth. Photosynthetically fixed C deposited at the soil surface can be delivered to deep soil horizons directly by root death; indirectly by biological or physical mixing; or by advection with soil solution (Strahm et al., 2009). The advective transport of dissolved or colloidal C is greatly facilitated by preferential flow channels, which reduce the travel time and potential for degradation during transport.

Hawaiian rainforest ecosystems lose mobile organic C from surface horizons to deeper ones and to streams creating pathways for removal of organically bound nutrients from terrestrial ecosystems (Hedin et al., 2003; Neff et al., 2000; Vitousek, 2004). The volcanic soils in these forests are rich in SRO minerals, which along with their strong C sorption capabilities also are prone to shrinking and swelling depending on rainfall frequency. As a consequence, cracks between peds provide preferential flow paths that carry organic matter from O and Bh horizons to deeper horizons where they are sorbed on SRO minerals (Marin-Spiotta et al., 2011). Marin-Spiotta et al. (2011) suggest both dissolved and particulate C are important vehicles for C transport in these soils, but they report only soluble C and do not offer a mechanism for the generation of mobilized organic carbon (MOC). The soils studied by Marin-Spiotta et al. (2011) receive about 3000 mm rainfall annually and experience periods of episaturation and anoxia that promote Fe reduction and lead to a net loss of Fe from the upper horizons. Reductive dissolution of Fe^{III}-oxides can promote colloid dispersion by dissolving the connective Fe cement holding aggregates together (Goldberg and Glaubig, 1987). Also, because Fe reduction consumes protons and causes an increase in solution pH (Gillespie, 1920; Ponnampuruma et al., 1966; Vesparaskas and Faulkner, 2001), it can indirectly influence colloid dispersion through development of negative charge on colloid surfaces (Bunn et al., 2002; Ryan and Gschwend, 1994). Therefore, Fe

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reduction can influence colloidal mobilization physically via dissolution of particles or chemically through changes in pH and corresponding particle surface charges (Thompson et al., 2006a). Fimmen et al. (2008) and Grybos et al. (2009) have suggested that organic matter is likely to disperse during Fe–C redox cycles, although they do not provide direct evidence of organic colloids.

Our research objective is to quantify changes in dissolved (<10 kDa), nanoparticulate (2.3–60 nm), and colloidal (60–430 nm) organic matter following Fe reduction events and induced pH increases. We use surface soils from Hawaiian rainforests and subject them to laboratory shifts in redox and pH to simulate conditions prevalent in these high-rainfall (>2500 mm/yr) ecosystems. We hypothesize that anoxic conditions destabilize C in organic-rich basaltic soils and generate mobile organic colloids. Thompson et al. (2006a) have shown that Fe reduction in similar-age Hawaiian soils promoted colloid dispersion primarily through the indirect effects of hydroxide production. We suspect that increases in pH associated with reduction reactions play a role in enhancing carbon release as well. To test these hypotheses, we measured the C content across three particle size classes following laboratory incubations of the soils exposed to either a 21-day period of anoxia or short-term pH adjustments.

2. Materials and methods

2.1. Site description

For this study, we selected surface mineral soil horizons from three sites on the Island of Hawai'i. The soils were collected in July 2009 from the Thurston (A horizon) and Laupāhoehoe (Ag/Bh horizon) sites reported in Kramer et al. (2012), and Pu'u Eke (Bh horizon) site reported in Marin-Spiotta et al. (2011). The Pu'u Eke soil (a Hydric Hapludand or Hydric Placudand) developed in the Pololu flows on Kohala Mountain, which are ca. 350 ka; the Laupāhoehoe soil (a Thaptic Udivitrand) formed in volcanic ejecta from Mauna Kea and is ca. 20 ka; and the Thurston soil (a Lithic Hapludand) lies on Kiluaea and formed in volcanic ejecta that is ca. 0.3 ka (Fig. 1). Detailed

characteristics of these soils are provided elsewhere (Kramer et al., 2012; Marin-Spiotta et al., 2011). Although SRO abundance increases progressively with age across these soil profiles as a whole, the SRO content of these surface mineral horizons peaks at the Laupāhoehoe site and then decreases in the Pu'u Eke site (Table 1). Once collected, the soils were stored at 4 °C under oxic conditions in the dark until used in the May 2010 experiments.

2.2. Anoxic experiment

Triplicate incubations of field-moist surface mineral soil horizons were suspended in 2.0 mM KCl at a soil:solution ratio of 1:10 (dry mass equivalent) in 12-mL polypropylene tubes. Soil-dry mass was determined after drying a separate subsample for 24 h at 110 °C. The suspensions were shaken in 12-mL polypropylene vials for 2 h on a horizontal shaker (~120 revolutions per minute, rpm). Three, 1.5-mL aliquots were removed and subjected to differential centrifugation targeting <430 nm and <60 nm particle-size fractions, as well as an ultrafiltration step (10 kDa) targeting a <2.3 nm fraction. The 430 nm cutoff was designed to target select particles below 0.45 μm, which is the cutoff for dissolved organic carbon used in the literature. The 60 nm value was selected to approximate conservatively a cutoff between nanoparticles (defined as <100 nm) and colloids (defined >100 nm). The <2.3 nm was chosen to represent a truly dissolved (non-particle) size fraction. These aliquots provided a pre-reaction sample set. The remaining suspension was opened in an anoxic chamber (95% N₂, 5% H₂) and allowed to equilibrate for 16 h before resealing. Suspensions were then secured on an end-over-end shaker (8 rpm) inside the glovebox and reacted for 21 d at room temperature (~25 °C). We monitored Fe^{II} production in the aqueous phase and terminated the experiment once Fe^{II} production began to increase substantially. Storing tropical soils at 4 °C has been recently shown to slow the recovery of Fe reducing organisms (Ginn et al., 2014) and that may have contributed to the slow emergence of aqueous Fe^{II}.

Following incubation, size-fractionated samples were isolated by differential centrifugation in an Eppendorf 5430 centrifuge with a F45-

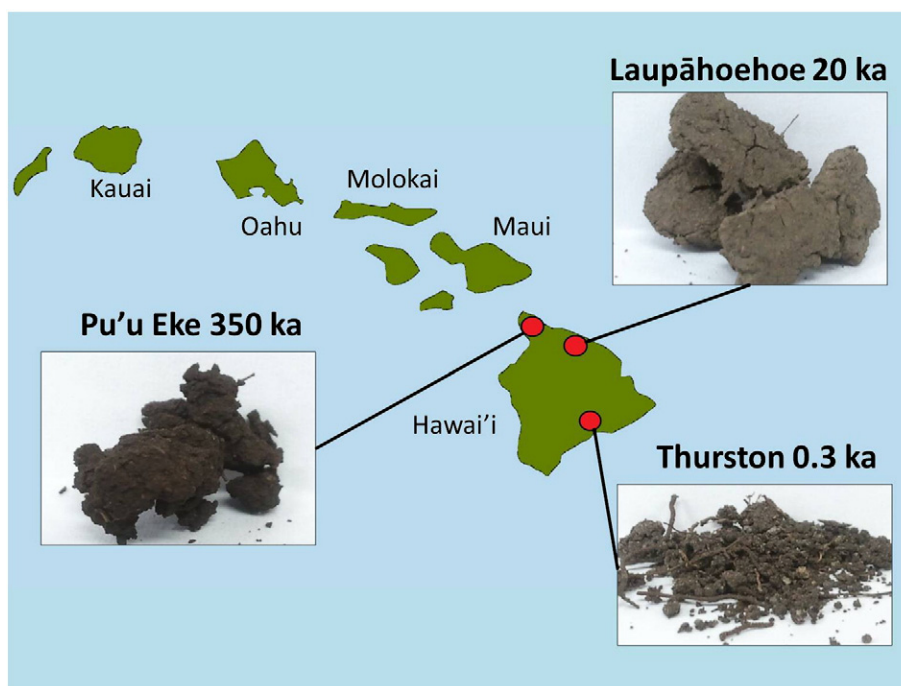


Fig. 1. Location of the experimental soils along with their approximate age and photo. All soils were collected from the volcanic Island of Hawai'i in July 2009. Thurston (A horizon) and Laupāhoehoe (Ag/Bh horizon) sites reported in Kramer et al. (2012), and Pu'u Eke (Bh horizon) site reported Marin-Spiotta et al. (2011). Pu'u Eke and Laupāhoehoe soils were visually more homogeneous and clayey than the Thurston soil (inset photos).

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