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Organic C and N stabilization in a forest soil: Evidence from sequential density fractionation

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Abstract

In mineral soil, organic matter (OM) accumulates mainly on and around surfaces of silt- and clay-size particles. When fractionated according to particle density, C and N concentration (per g fraction) and C/N of these soil organo-mineral particles decrease with increasing particle density across soils of widely divergent texture, mineralogy, location, and management. The variation in particle density is explained potentially by two factors: (1) a decrease in the mass ratio of organic to mineral phase of these particles, and (2) variations in density of the mineral phase. The first explanation implies that the thickness of the organic accumulations decreases with increasing particle density. The decrease in C/N can be explained at least partially by especially stable sorption of nitrogenous N-containing compounds (amine, amide, and pyrrole) directly to mineral surfaces, a phenomenon well documented both empirically and theoretically. These peptidic compounds, along with ligand-exchanged carboxylic compounds, could then form a stable inner organic layer onto which other organics could sorb more readily than onto the unconditioned mineral surfaces ("onion" layering model).

To explore mechanisms underlying this trend in C concentration and C/N with particle density, we sequentially density fractionated an Oregon andic soil at 1.65, 1.85, 2.00, 2.28, and $2.55 \,\mathrm{g \, cm^{-3}}$ and analyzed the six fractions for measures of organic matter and mineral phase properties.

All measures of OM composition showed either: (1) a monotonic change with density, or (2) a monotonic change across the lightest fractions, then little change over the heaviest fractions. Total C, N, and lignin phenol concentration all decreased monotonically with increasing density, and ¹⁴C mean residence time (MRT) increased with particle density from ca. 150 years to >980 years in the four organo-mineral fractions. In contrast, C/N, ¹³C and ¹⁵N concentration all showed the second pattern. All these data are consistent with a general pattern of an increase in extent of microbial processing with increasing organo-mineral particle density, and also with an "onion" layering model.

X-ray diffraction before and after separation of magnetic materials showed that the sequential density fractionation (SDF) isolated pools of differing mineralogy, with layer-silicate clays dominating in two of the intermediate fractions and primary minerals in the heaviest two fractions. There was no indication that these differences in mineralogy controlled the differences in density of the organomineral particles in this soil. Thus, our data are consistent with the hypothesis that variation in particle density reflects variation in thickness of the organic accumulations and with an "onion" layering model for organic matter accumulation on mineral surfaces. However, the mineralogy differences among fractions made it difficult to test either the layer-thickness or "onion" layering models with this soil. Although SDF isolated pools of distinct mineralogy and organic-matter composition, more work will be needed to understand mechanisms relating the two factors.

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Keywords: Soil organic matter; Particle density; ¹³C; ¹⁵N; ¹⁴C; MRT; Lignin; Hydroxyfatty acids; Cutin; Suberin; X-ray diffraction; Glycoprotein; Amino sugar; Protein; Amino acid; Smectite; Magnetite; Acid/aldehyde ratio; Carbon; Nitrogen; Sulfur; Soil C stabilization

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1. Introduction

Understanding controls on soil organic matter (SOM) storage is critical to quantifying changing rates of C cycling and storage in response to global climate change. Additionally, SOM plays critical roles in governing agricultural and forest soil dynamics and ecosystem productivity. Soil organic matter (OM) dynamics are complex, requiring understanding of biological and chemical interactions between organic compounds and mineral soil constituents. In mineral soil, OM accumulates mainly on and around surfaces of silt- and clav-size particles. When fractionated according to density, C and N concentration (per g fraction) and C/N of these soil organo-mineral particles decrease with increasing particle density across soils of widely divergent texture, mineralogy, location, and management (Fig. 1). Portions of this trend in C/N are easily understood. The lightest fractions $(<1.6-1.8 \,\mathrm{g \, cm^{-3}})$ have high C/N due simply to the high content of plant-derived polysaccharides, lignin, cutin, suberin, and charcoal. The continued decrease in C/N above densities $> 2.0 \,\mathrm{g \, cm^{-3}}$ was commented on by Oades (1989) but remains unexplained.

To understand this trend in C/N, it is helpful to replot the data underlying Fig. 1 to show particle density as a function of C concentration (Fig. 2). The strong inverse relation suggests that much of the variation in density of the soil particles is due to differences in C concentration. Alternatively, some could also be due to variation in density of the mineral or organic phases. The first explanation, however, implies that the thickness of the organic accumulations decreases with increasing particle density. Thus it may be reasonable to recast the trend in C/N with density as reflecting a decrease in C/N with decreasing thickness of the organic layer accumulated on mineral surfaces. Layering, perhaps, is not the best term to describe this phenomenon, as there is ample evidence that the OM is distributed discontinuously over the mineral surfaces at scales ≪1µm (Mayer, 1999; Kaiser and Guggenberger, 2003; Zimmerman et al., 2004). However, scanning electron microscopy at scales $> 1 \,\mu m$ shows very few clean mineral surfaces in the A horizons of most mineral soils (e.g., Spycher et al., 1986), thus we use "layer" here for convenience.

A low C/N in thin organic layers on mineral surfaces can be explained at least partially by especially stable sorption of nitrogenous compounds (glycoproteins, amino sugars, and proteins) directly to mineral surfaces, a phenomenon well documented both empirically and theoretically. Much soil organic N occurs as protein (Schulten and Schnitzer, 1998; Knicker, 2000; Knicker et al., 2000; DiCosty et al., 2003; Martens and Loeffelmann, 2003; Schmidt-Rohr et al., 2004). A significant portion may occur as heterocyclic N (Mertz et al., 2005; Smernik and Baldock, 2005), which can be biotic (e.g., nucleic acids), combustion products associated with charcoal, or fossil organics inherited from



Fig. 1. C:N ratio vs. particle density for soils worldwide and the andic soil studied here. • Andic Dystrudept (this study); • Marine sediment (Arnarson and Keil, 2001); • Haploxerolls, 3ky unit (Baisden et al., 2002); • Haploxeralfs, 200ky unit (Baisden et al., 2002); • Alfisols, 600ky unit (Baisden et al., 2002); • Alfisols, 3my unit (Baisden et al., 2002); • Chromustert,Qld (Golchin et al., 1994a, b); • Pellustert (Golchin et al., 1994a, b); • Rhodoxeralf (Golchin et al., 1994a, b); • Pellustert (Golchin et al., 1994a, b); • Andisol, grassland (Golchin et al., 1997); • Andisol, afforested (Golchin et al., 1997); • Andisol, forest (Golchin et al., 1997); • Xeropsamment, Mudflow A (Sollins et al., 1983); • Xeropsamment, Mudflow B (Sollins et al., 1983); • Xeropsamment, Mudflow C (Sollins et al., 1983); • Xeropsamment, Mudflow B (Sollins et al., 1983); • Xeropsamment, Mudflow C (Sollins et al., 1983); • Andisol (Sollins et al., 1983); • Andisol, 500 µm (Turchenek and Oades, 1979); • Haploboroll, 200 µm (Turchenek and Oades, 1979); • Haploboroll, 5-2 µm (Turchenek and Oades, 1979); • Haploboroll, 200 µm (Turchenek and Oades, 1979); • Haploboroll, <0.4 µm (Turchenek and Oades, 1979); • Argixeroll, clay-size (Young and Spycher, 1979); • Sideraquod, clay-size (Young and Spycher, 1979); • Dystrandept (Young and Spycher, 1979); • Argixeroll, clay-size (Young and Spycher, 1979).

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